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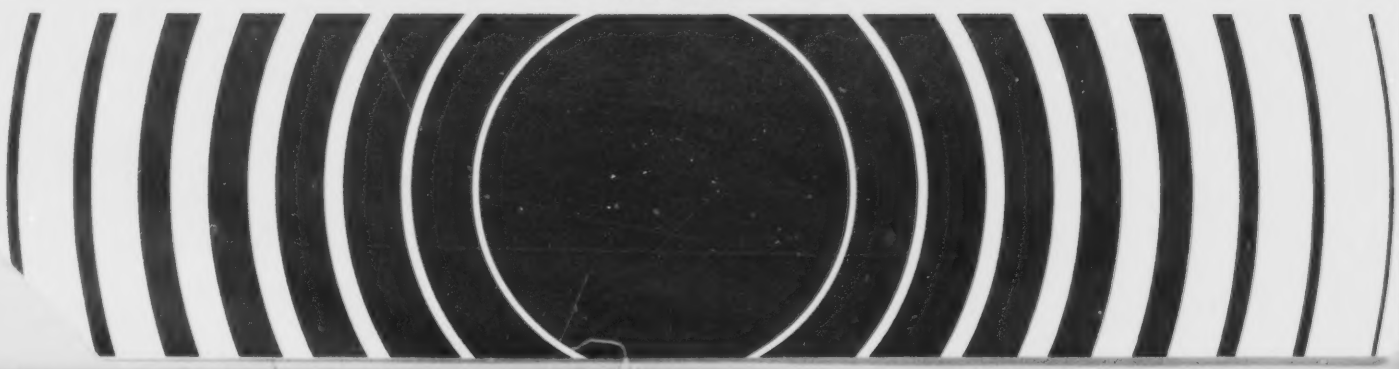

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A SURVEY OF THE AVAILABLE METHODS OF SOLIDIFICATION FOR RADIOACTIVE WASTES

by

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Office of Radiation Programs
U.S. Environmental Protection Agency
Washington, D.C. 20460

November 1978

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PREFACE

The Office of Radiation Programs of the U. S. Environmental Protection Agency carries out a national program designed to evaluate population exposure to radiation, and to promote development of controls necessary to protect the environment and public health. The mission of the Technology Assessment Division is to provide the primary assessments of the technologies currently utilized, or being proposed which have a potential radiation impact on man or his environment. This report was prepared as a survey and technology assessment of the currently available and proposed methods of solidification for radioactive wastes.

Readers of this report are encouraged to inform the Office of Radiation Programs of any omissions or errors. Comments or requests for futher information are also invited.

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Director
Technology Assessment Division (ANR-459)
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ABSTRACT

This report reviews the numerous solidification techniques and related matrix materials that are presently being offered or proposed for incorporating the radionuclides into an immobile material. Both high- and low-level waste solidification processes are covered. Key features of the equipment used in individual solidification processes are discribed. At present the high-level waste solidification methods are being developed by the Government, while the low-level waste solidification methods are being developed commercially.

TABLE OF CONTENTS

EPA Review Notice	ii
Preface	iii
Abstract	iv
Table of Contents	v
List of Tables	vii
List of Figures	viii
I. Introduction	1
II. Background	2
1.0 Solidification	3
2.0 Waste Classification	4
3.0 Cited References	5
III. High-Level Radioactive Waste Solidification Techniques	6
1.0 Calcination - Solidification Processes	6
1.1 Fluidized-Bed calcination	6
1.2 Spray Calcination	9
1.3 Pot Calcination	9
1.4 Rotary Kiln Calcination	12
2.0 Cement Solidification	12
3.0 Glassification - Solidification Processes	14
3.1 In-Can Melting	14
3.2 Continuous Melting	15
4.0 Other Solidification Processes	18
5.0 Cited References	18
6.0 Additional References	22

IV. Low-Level Radioactive Waste Solidification Techniques 30

1.0 Solidification of Low-Level Radioactive Wastes in Bitumen 33

1.1 Stirred - Evaporator Batch Process 35
1.2 Emulsified Bitumen Process 35
1.3 Film Evaporation Process 38
1.4 Screw Extrusion Process 40
1.5 Other Research 42
1.6 Advantages and Disadvantages 42

2.0 Solidification of Low Level Radioactive Wastes in Cement 44

2.1 In-drum Mixer Process 49
2.2 External Mixer Process 52
2.3 Other Processes 54
2.4 Advantages and Disadvantages 54

3.0 Polymeric Solidification Processes for Low Level Radioactive Wastes 56

3.1 Urea Formaldehyde Method 56
3.2 Advantages and Disadvantages 61
3.3 Other Polymeric Processes 62

4.0 Use of Absorbents for Solidification of Low-Level Radioactive Wastes 64

5.0 Cited References 64

6.0 Additional References 70

V. Leaching Studies 72

1.0 References 74

VI. Conclusions 76

Appendix A - EPA Presentation Before National Academy of Sciences, February 1, 1977. 78

Appendix B - President Carter's Statement on Nuclear Power Policy, April 7, 1977. 82

Appendix C - Status of Fuel Reprocessing in the U. S. 85

LIST OF TABLES

I.	Solidification Agents and Vendors	31
II.	Cementation Practices at Various Establishments	47
III.	Comparisons of Leach Rates for Various Solidified Waste Products	73

LIST OF FIGURES

1.	Fluidized-Bed Calciner	8
2.	Spray Calciner	10
3.	Pot Calciner	11
4.	Rotary Kiln Calciner	13
5.	In-Can Melter	16
6.	Continuous Melter	17
7.	Low-Level Waste Processing Steps	34
8.	Stirred-Evaporator Batch Process	36
9.	Emulsified Bitumen Process	37
10.	Turbulent-Film Evaporation Process	39
11.	Screw Extruder Evaporation Process	41
12.	Flow Diagram For Cement Incorporation Processes	50
13.	Example of In-Drum Mixing Process	51
14.	Example of External Mixing Process	53
15.	Flow Diagram for UF Incorporation Processes	57
16.	Urea Formaldehyde Incorporation Process - External Mixing	58

I. INTRODUCTION

It is the intent of this report to review the various solidification systems and matrix materials that are presently employed, proposed or under development; to include operational descriptions, where possible, and also to indicate how these materials and methods are affected by characteristics of the wastes. Both high - and low-level waste solidification processes are reviewed and described.

This review is being done in accordance with the Environmental Protection Agency's mission of assessment of the technologies currently being utilized, or proposed for use in activities which may have a potential radiation impact on man or his environment. The EPA has for many years been involved in radioactive waste management activities, and in particular the waste characteristics which include inventories, sources and waste forms. The Office of Radiation Programs have undertaken many in-house efforts to address these problems and also to interact with other agencies and groups to effect efficient solutions to radioactive waste disposal problems. Appendix A is the presentation EPA made before the National Academy of Sciences on February 1, 1977 concerning solidification.

II. BACKGROUND

The nuclear power and radioisotope industry that has developed in this century has presented a challenging problem in the management of radioactive waste materials. The treatment and disposal of radioactive wastes involve both environmental considerations and technical processing problems which are complex and potentially far-reaching because of the long effective half-lives of certain radionuclides. Waste management as used here encompasses the treatment and packaging as well as the disposal (i.e., burial) of the wastes. Unlike many industrial wastes, radioactive wastes are not susceptible to neutralization techniques. Natural decay is the only means of destroying radioactivity (the process of transmutation is not considered viable at this time). Each radionuclide decays at its own particular rate regardless of outside influences. Since the various waste radionuclides have decay rates ranging from days to thousands of years, treatment and processing such as solidification techniques, become an important factor in radioactive waste management. As it is virtually impossible with present technology to destroy the radioactivity associated with waste materials, containment or dispersal have been the only two ways of dealing with these unwanted products (1).

The Environmental Protection Agency (EPA) considers a high degree of permanent containment of radioactive materials to be the primary requirement for environmentally adequate radioactive waste management; i.e., the assurance of isolation of the waste from the environment (biosphere) for the duration of the wastes hazardous lifetime (2). As a result EPA promotes the policy of containment rather than the concept of planned release or dispersion as currently used in the regulation of gaseous emissions from nuclear facilities (3).

EPA recognizes the need for environmental criteria and standards for waste management. The Agency was directed to establish environmental radiation protection standards for the disposal of high-level radioactive waste as a result of the Presidential Nuclear Policy Statement of October, 1976 (4). In implementing EPA's program for preparing environmental criteria and establishing generally applicable radioactive waste disposal standards to protect the environment and public health the Agency's programs center on several areas, one of which is the assessment of the performance of engineering barriers such as solidified waste matrices. In reviewing the available information EPA has relied on both the available and future technologies developed by the Department of Energy and the commerical industry.

1.0 Solidification

The solidification process has become an important step in the scheme of waste management and the philosophy of environmental containment. This solidification step converts the waste, usually a liquid or semi-liquid, into a physical form which can be handled, stored and disposed of more safely and conveniently. It also reduces the volume of the waste by removing liquids; and reduces the potential for movement of the incorporated radionuclides through the geosphere after disposal.

There are many varieties of solidification materials and techniques available. In all cases the method of solidification used should not be a reversible process which can allow the solid to return to a liquid form. Estimation of the rate of leaching from a solidified matrix during disposal is one of the important considerations in the assessment of a solidification method as it will strongly influence the amount of treatment, containment, and surveillance that will be needed. Low matrix solubility will improve the safety of waste management implemented through isolation by reducing further the likelihood of an unplanned release.

In any solidification or stabilization process where radioactive materials are used, the process and operational conditions are complicated by the difficulties of remotely operating and maintaining equipment. Attention must be given to reliability, and rugged equipment may have advantages over complex or sensitive equipment.

Present regulations require that commercially-generated (as opposed to those generated by the Federal Government) high-level liquid radioactive wastes be solidified within five years and shipped to a Federal repository within ten years (5). The necessity of solidifying the low-level wastes is not the result of burial considerations, but of transportation considerations. The transport of radioactive wastes is done in compliance with regulations issued by the Department of Transportation and by the Nuclear Regulatory Commission for the safe transport of radioactive materials (6, 7).

2.0 Waste Classification

There are two classes of radioactive wastes for which solidification techniques have been developed: high-level and low-level.

The high-level, high-activity radioactive wastes are those liquid wastes, or the solidified products of those wastes, which are associated with the first cycle solvent extraction or equivalent in a facility for processing irradiated reactor fuels. Research and development on solidification processes have been underway for high-level wastes since the late 1950's. A number of methods for processing these wastes have been proposed and investigated. These include conversion of the liquid wastes to a stable calcined powder or granules, and conversion of the calcine or liquids to a glass form. Other methods include fixation within solids such as clays, cement, ceramics or similar materials.

The low-level radioactive wastes include those radioactively contaminated wastes generated from the nuclear fuel cycle operations and facilities not specifically designated high-level. The majority of these wastes are generated by nuclear power plants and are usually in the form (prior to solidification) of processed waste water, evaporator concentrates, sludges, filter-aids, demineralizer resins, miscellaneous scrap material, etc. These wastes encompass a broad spectrum of materials varying widely in chemical and radioactive content. Various solidification techniques for these wastes have been demonstrated in operating nuclear facilities throughout the world for many years. The various solidification agents include portland cement, concrete, plaster of paris, asphalt (or bitumen), polymers, and a blend of absorbent material and cement or plaster.

It should be noted throughout the following sections that the solidifcation processes and techniques developed for the two classes of wastes, (high and low-level), are entirely different. The major reasons are that the high-level wastes have higher thermal activity, larger quantities of radioacitivty per volume, and higher radiation fields than the low-level wastes. As a result of these differences the techniques for high-level waste require entirely remote operations and much greater shielding considerations than the low-level waste techniques; although, in both cases there are common considerations in the techniques for assessing the potential environmental impact from the solidified waste form. They include such properties as: (a) leachability; (b) thermal conductivity; (c) chemical stability; (d) radiation resistence; and (e) mechanical ruggedness.

3.0 Cited References

1. P.N. Cheremisinoff and W.F. Holcomb, "Management of Hazardous and Toxic Wastes," Pollution Engineering, Vol. 8, No. 4, page 24, April 1976

2. W. D. Rowe and W. F. Holcomb, "The Hidden Commitment of Nuclear Wastes," Nuclear Technology, Vol. 24, No. 3, page 286, December 1974.

3. Statement of David G. Hawkins, Assistant Administrator for Air and Waste Management, Environmental Protection Agency, before the Subcommittee on Nuclear Regulation of the Committee on Environmental and Public Works, U.S. Senate, March 22, 1978.

4. "The White House Fact Sheet -- President's Nuclear Waste Management Plan," Office of the White House Press Secretary, Washington, D.C., October 27, 1976.

5. "Licensing of Production and Utilization Facilities -- Policy Relating to the Siting of Fuel Reprocessing Plants and Related Waste Management Faciliites," Code of Federal Regulation, Title 10, Chap. I., Part 50, Appendix F, U.S. Government Printing Office, Washington, D.C., 1976.

6. "Shipper - General Requirements for Shipments and Packaging," Code of Federal Regulations, Title 49, Chap. I, Part 173, U.S. Government Printing Office, Washington, D.C., 1976.

7. "Packaging of Radioacitve Material for Transport and Transportation of Radioactive Material Under Certain Conditions," Code of Federal Regulations, Title 10, Chap. I, Part 71, U.S. Government Printing Office, Washington, D.C. 1976.

III. HIGH-LEVEL RADIOACTIVE WASTE SOLIDIFICATION TECHNIQUES

High-level liquid radioactive wastes are produced during spent-fuel recovery operations at reprocessing plants. These wastes are the highly radioactive solutions that remain after the separation of the unused (or unfissioned) uranium from the fission product waste produced during reactor operation. Reprocessing of spent fuels from the commercial nuclear power industry is not being done in the United States at present (1). See Appendix B for text of statement. However, a substantial amount of these wastes are on hand from past reprocessing of defense and commercial materials (2-10). See Appendix C for status of fuel reprocessing.

Extensive research and development has taken place throughout the world for the past 20 years on possible alternatives for processing these high-activity wastes (11-14). Many of these techniques have been successfully demonstrated on either pilot-plant or full scale operation. These alternatives have resulted in a number of solidification technologies, of which two seem to have emerged as the most prominent - calcination and glassification. While both solidification technologies are discussed as possible alternatives it is expected that they will complement one another by using the calcination step as a conversion process from liquid to solid (with volume reduction included) and the glassification step will provide a vitrified solid for terminal disposal operations.

1.0 Calcination-Solidification Processes

Calcination is a process that heats a material to a temperature which will drive off most volatile material, but not hot enough to cause fusing of the material. Four major U. S. and foreign technologies relate to the calcination process; they are fluidized-bed, spray, pot, and rotary kiln calcination (15-20).

1.1 Fluidized-Bed Calcination

The fluidized bed calcination process was the earliest technique investigated for the conversion of radioactive waste solutions to solids (15, 21). Development of this process was sponsored under an Atomic Energy Commission (now the Department of Energy) program which started in 1955. A Waste Calcining Facility (WCF) was built at the Idaho National Engineering Laboratory near Idaho Falls, Idaho, as part

of the Idaho Chemical Processing Plant. It is the world's only plant scale unit which converts high-level radioactive liquids to solids using fluidized bed calcination (3, 11, 14, 15, 21-24). From 1963 through 1977 the WCF converted about 12 thousand cubic meters of high-level radioactive liquid wastes to about 1500 cubic meters of granular calcine solids (5). Solidification of the nitric acid fuel dissolution wastes began at a net rate of 227 liters per hour and has been increased to about 340 liters per hour. Wastes from sulfuric acid and hydrofluoric acid fuel dissolutions also have been solidified.

The liquid waste is pneumatically atomized and sprayed into a fluidized bed of granular solids operating at a temperature of 400-500 C. Here the liquids are vaporized and the gaseous materials enter an off-gas system, while the metallic salts are converted to their corresponding oxides or fluorides and deposited layerwise on the spherical bed particles. The solid particles are removed continuously from the fluidized bed and transported pneumatically to storage facilities. The waste solutions calcine to such compounds as Al_2O_3, CaF_2, ZrO_2, Fe_2O_3, $Al_2(SO_4)_3$, etc and also certain amounts of amorphous materials; while water vapor, oxides of nitrogen, and other volatiles leave with the calciner off-gas. During calcination of hydrofluoric acid-type waste, the feed also includes the additon of calcium nitrate to complex the fluoride ion thus preventing the formation of corrosive fluoride gases.

The volumetric ratio of atomizing air to liquid feed in the waste nozzle is varied as necessary to cause sufficient breakup of bed particles to control the average bed particle size at a desired value. Heat for the endothermic calcination process is supplied by in-bed combustion of kerosene injected and atomized by oxygen through fuel nozzles located in the walls of the calciner vessel (22, 23). Startup of the process requires heating the fluidized bed to greater than 360 C using heated fluidizing air. The liquid wastes containing nitrate are then injected through a waste atomizing nozzle, followed immediately by the injection of the fuel-oxygen mixture through the fuel atomizing nozzle. Ignition of the fuel-oxygen mixture is spontaneous at temperatures above 270 C in the presence of nitrates. Off-gas from the calciner is cleaned by successive passage through a dry cyclone, a wet venturi scrubbing system, silica gel absorbers, and high efficiency particulate filters to remove contaminants before the gas is exhausted to the atmosphere. Figure 1 shows a fluidized bed calciner as used at the Idaho Facility.

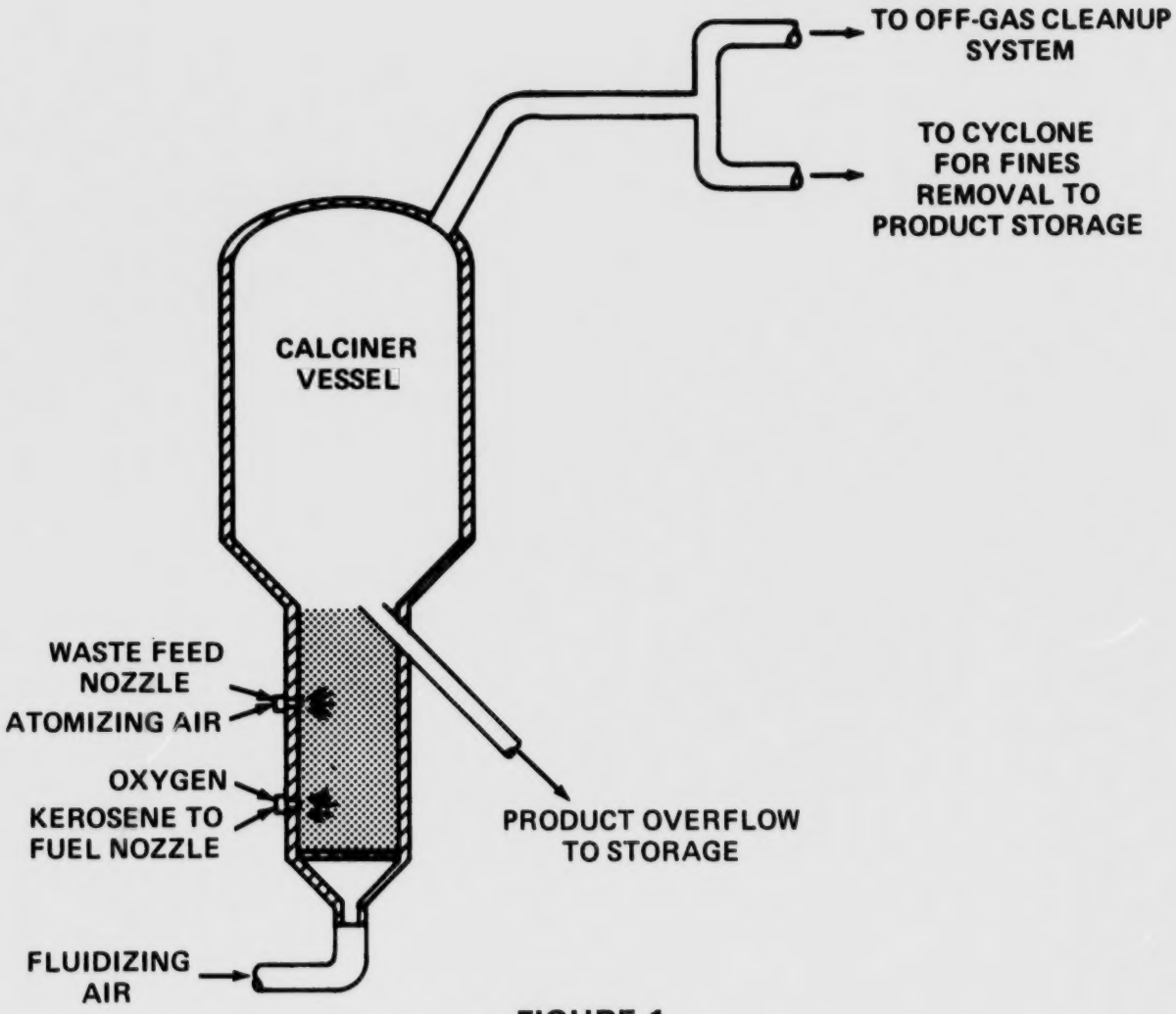

FIGURE 1

FLUIDIZED-BED CALCINER

1.2 Spray Calcination

This process has been under development at the Department of Energy's Hanford Reservation, Richland, Washington, for over 15 years and is presently undergoing tests by the Battelle Pacific Northwest Laboratories using simulated wastes (11, 15, 18-20, 25).

The liquid waste is fed to a pneumatic air atomizing nozzle in the top of a heated (700-800 C) spray calciner vessel. Convective and radiant heat transfer flash dries and calcines the atomized droplets as they fall through the chamber. Over half the calcine falls into an outlet cone while the balance collects as a coating on sintered stainless steel filters located in the vessel gas outlet. Periodically the filter coating is blown back to dislodge the powder which falls into the cone. Figure 2 illustrates a spray calciner vessel. The material in the cone is discharged to either canisters or into a close-coupled melter. The process has operated at feed rates from 75 to 300 liters per hour. The gaseous and liquid effluents can be treated by the usual off-gas purification type equipment, i.e., wet scrubbers, quenching spray tower, venturi scrubber, cyclone, condenser, knockout pot, silica gel sorption towers etc. A variation of the spray calcining process has been under devlopment and in use at the Karlsruhe (W. Germany) Nuclear Research Center since the early 1970s (26, 27).

1.3 Pot Calcination

This process was developed at the Department of Energy's Oak Ridge National Laboratory and was demonstrated at the Hanford Reservation facilities (11, 15, 18, 19, 25).

The liquid waste is fed to a heated pot located in a multiple-zone heating and cooling furnace. The pot also serves as the final storage vessel. As the liquid waste concentrates, it begins to form a scale on the inside walls of the pot. As calcination continues, the scale grows in thickness while the feed rate decreases and finally is stopped. At this point, the scale has grown inward and upward to fill the pot, except for the cone-shaped void at the top. Heating is continued until the liquid is boiled to dryness and all the waste has been calcined at temperatures up to 900 C. The pot is then cooled, removed, sealed and stored. The total time cycle for filling, cooling and changeout of an 20 cm. dia pot with 183 cm of calcine is about 44 hours at an average processing rate of 11 liters per hour. Figure 3 shows the pot calciner.

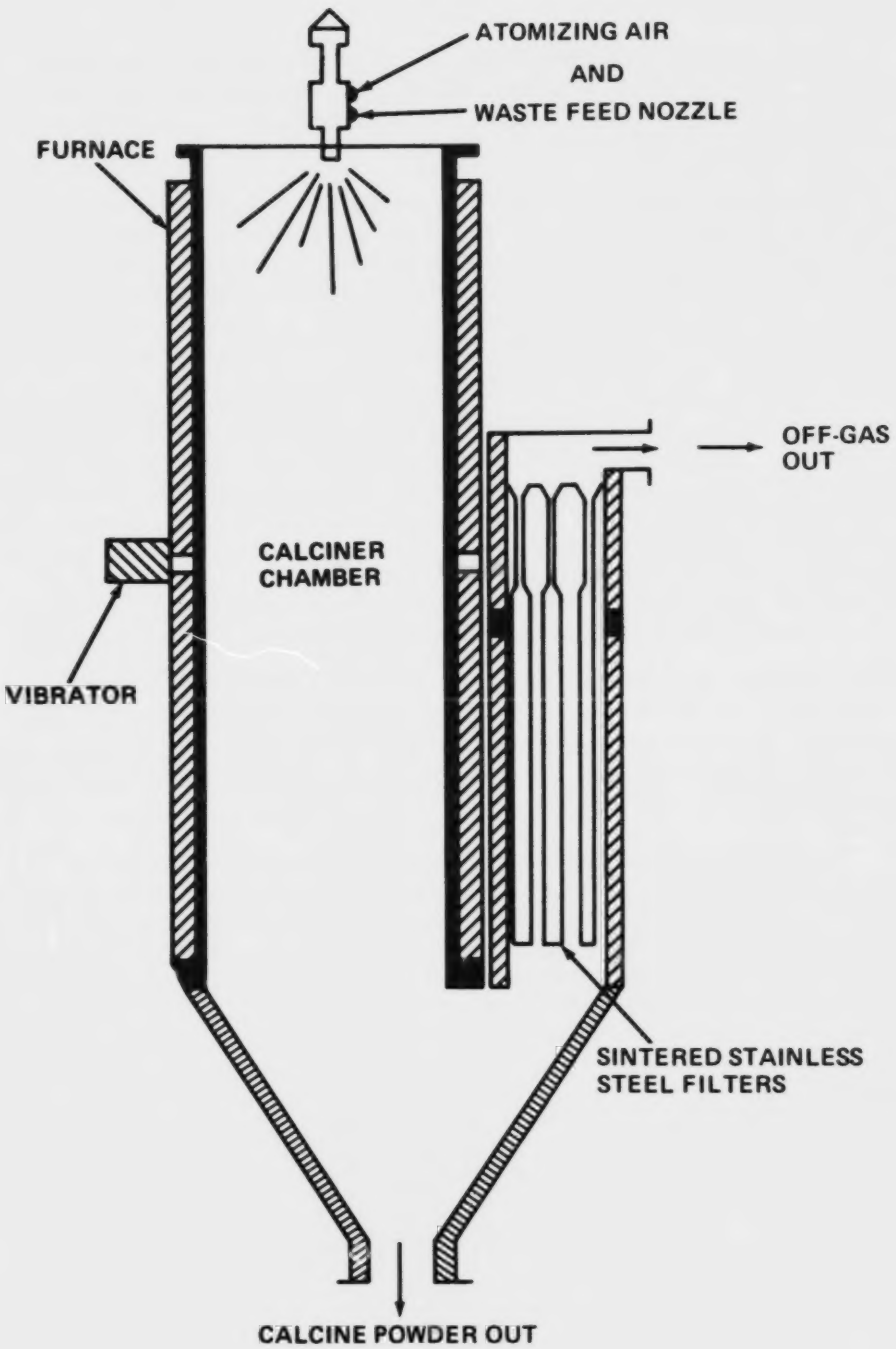

FIGURE 2

SPRAY CALCINER

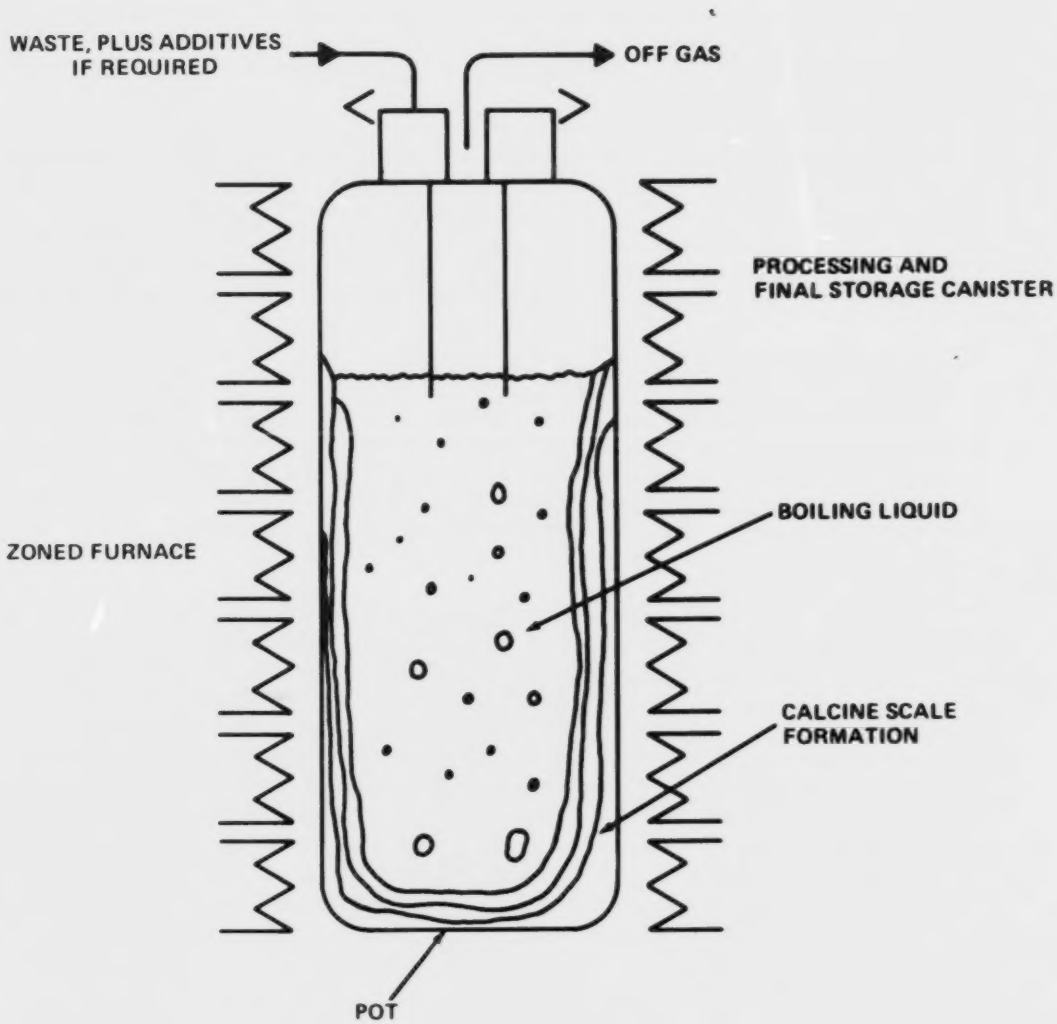

FIGURE 3

POT CALCINER

1.4 Rotary Kiln Calcination

This process was developed in the early '60s at the Department of Energy's Brookhaven National Laboratory. In the rotary-ball kiln calciner the waste solution is dripped or sprayed onto a bed of moving metal balls in a slowly rotating cylinder, electrically-heated to a temperature between 600 and 800 C. The product formed from deposition of the oxides of the metallic salts on the balls is pulverized during kiln operation. The capacity of a rotary-ball kiln calciner is based on the surface to bed heat transfer coefficient obtainable and effectively dispersing heat throughout the length of the calciner vessel. It is estimated that the capacity could be as high as 760-1130 liters per hour (11, 15, 20). Figure 4 illustrates a rotary kiln calciner.

The French Commissariat a l'Energie Atomique facility at Marcoule is using a variation of the rotary kiln process in connection with a continuous melter process for the conversion of high-level liquid wastes to a glass. The feed rate is expected to be 30-40 liters/hr (28). All recent development and commercialization activities concerning this process has been accomplished at the Marcoule facility.

2.0 Cement Solidification

The use of concrete as a matrix for solidification of the high-level radioactive wastes at DOE's Savannah River Plant (SRP) has been studied on an experimental, laboratory-scale program using both simulated and actual radioactive waste sludges (6, 29-33). Because of dilution and aging, the SRP high-level waste is suitable for fixation in cement.

The studies indicated that formulations of cement with washed and dried sludge powders are easy to prepare, and the waste is chemically compatible with concrete. Sludge loadings up to 40 wt% give waste forms with generally excellent properties. The concrete matrix properties studied were: (a) compressive strength; (b) cesium, strontium and alpha emitter leachability; (c) radiation and thermal stability; and (d) types of cement and sludge compositions used. The initial results indicated compressive strengths of 2000-3000 psi for formulations with 40% sludge content; reasonable long-term thermal and radiation stability; and strontuim $^{-90}$ and alpha emitter leachabilities typically in the 10^{-5} g/cm^2-d or greater range.

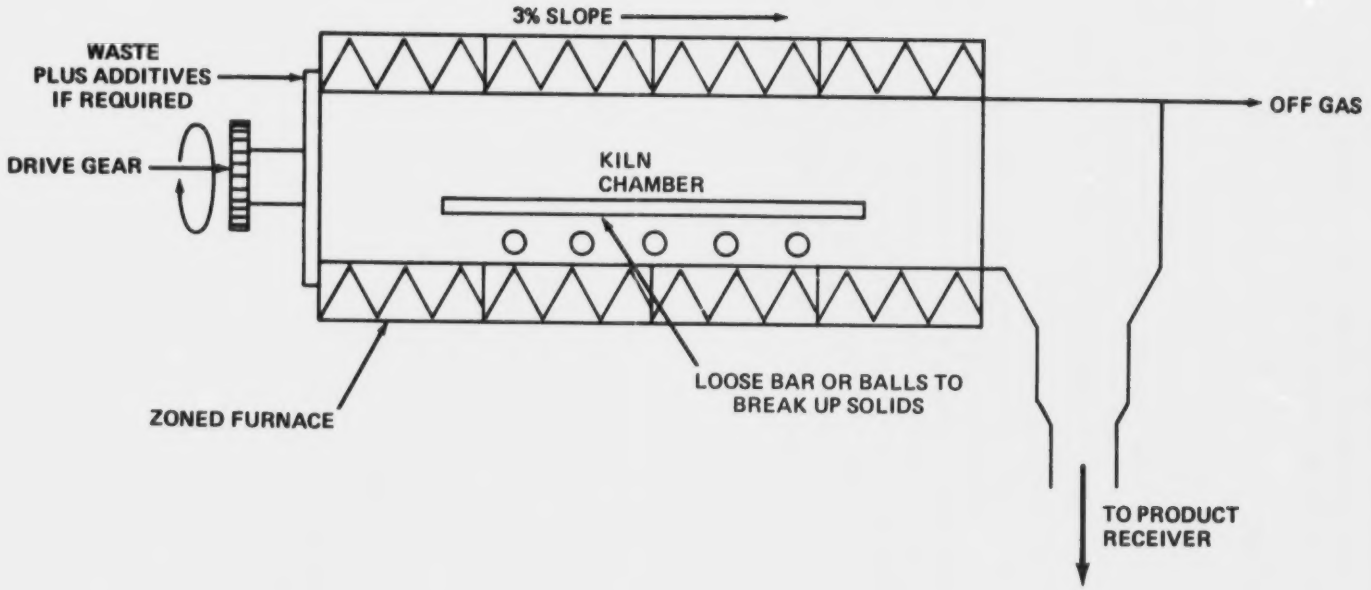

FIGURE 4

ROTARY KILN CALCINER

Some problems encountered were high cesium - 137 leachibility (less than 10^{-3} g/cm^2-d); steam generation if heated in closed containers, and excessive cement set times; although some methods were developed to overcome these problems.

3.0 Glassification - Solidification Processes

In connection with the calcination/solidification technique of immobilizing the high-level liquid radioactive wastes, the U.S. and the other countries using nuclear power have directed their research and development toward the goal of glassification (or use of similar vitreous forms) of these wastes for disposal in geological repositories.

The use of glass or glasslike materials provides a highly immobile solid form for fixing the wastes. In addition, it reduces air-borne releases, provides a less leachable material, is chemically, thermally, and radiolytically stable, and provides a suitable heat dissipating solid for geologic isolation. Borosilicate glass appears to be the preferred form in most glass programs. It has relatively low temperatures of formation, low corrosiveness to container material during formation, high retention of volatile species during processing, and low leach rates (11, 14, 16-18, 34). There are two processes which have been developed in the U.S. as candidates for commercial use: in-can melting and continuous melting.

3.1 In-Can Melting

The in-can melting batch process is being developed by Battelle's Pacific Northwest Laboratories for the Department of Energy. The process was developed as a tandem system coupled to the spray or fluidized bed calcination process.

The calcine powder falls directly into a close-coupled melter canister along with specially-formulated frit. The frit and the calcine are melted together in a metallic canister at processing temperatures of 1000-1100 C using a multizone furnace.

The advantages include: (a) minimum process steps and equipment; (b) non-transfer of melt; (c) everything entering the melter (except some volatile species) is fixed in the glass; (d) disposable melter; and (e) the melting step is flexible enough to accommodate a wide range

of calcine characteristics and compositions, i.e., it could also use the calcine product from other processes such as spray or rotary kiln calcination. Melting rates up to 100 kg/hr have been achieved using nonradioactive simulated high-level waste. Figure 5 illustrates the in-can melting process.

Even through the melting is a batch process and the calcination step is a continuous process, by using a diverter valve and multiple melting furnace-canisters, the systems are compatible (11, 19, 25, 35-38).

3.2 Continuous Melter

Battelle's Pacific Northwest Laboratories also have under development a continuous melter process (also called joule heated melter). This process is closely allied with the commercial electric glass melter processes and its flexibility allows it to be coupled to different kinds of waste calciners and even receive liquid wastes directly.

The system has a long life time while producing a high quality glass with low off-gas effluents. The process is carried out at temperatures ranging from 1000 to 1200 C in a refactory-lined melter with internal electrodes; the molten glass acts as its own electrical resistance heating element. Prototypes have operated at feed rates up to 100 kg/hr. The system also allows for flexibility in glass composition and for the controlled draining of glass from the melter which can permit changes in the final waste form package (11, 18, 25, 36, 38). Figure 6 shows the continuous melter.

The French are developing a system using the rotary calciner coupled to a melting furnace which is scheduled for full scale operation in 1978. In addition, the West German Government is developing systems using the spray calciner coupled to a continuous melter (27, 28).

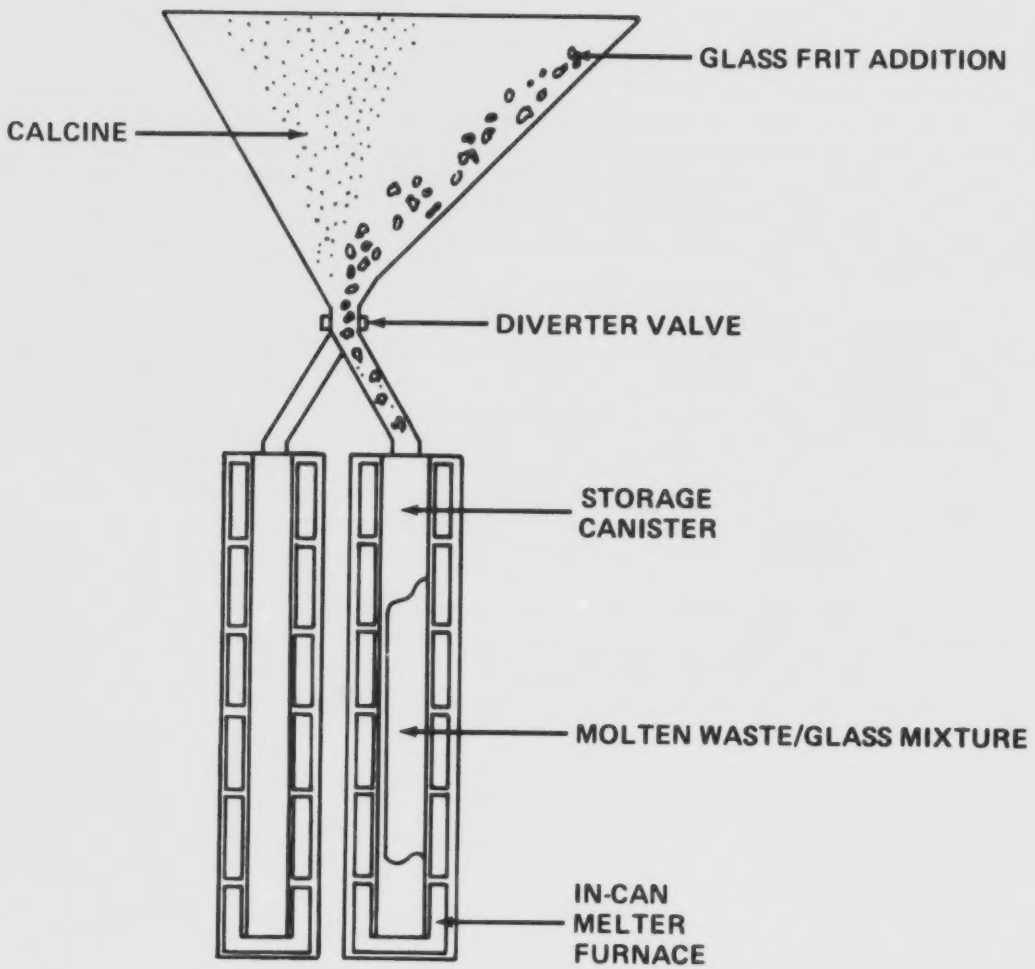

FIGURE 5

IN-CAN MELTER

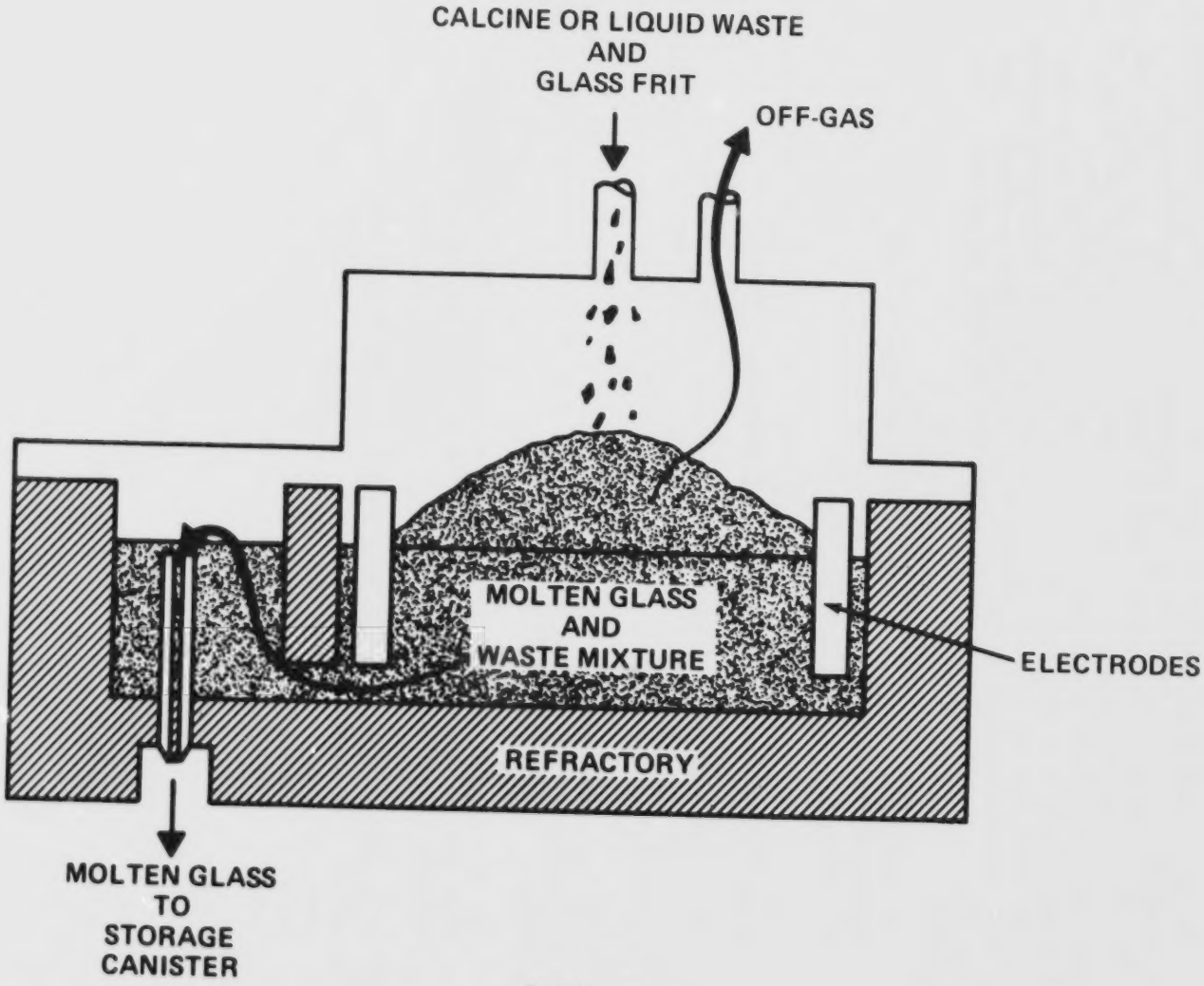

FIGURE 6

CONTINUOUS MELTER

4.0 Other Solidification Processes

There are numerous other processes and methods being investigated as alternatives to calcination and glassification to reduce costs, simplify processing operations, increase waste form compatibility, and to increase the reliability of the inertness and low dispersibility of waste form. Many of these processes are under laboratory-scale development while others are variations of the present techniques (3, 11, 13, 14). The various alternatives have not been under development as long as the existing glassification and calcination processes, and thus the best combinations and options have not been optimized. They include:

(1) Supercalcine - a process to produce an assemblage of thermodynamically stable crystalline ceramic waste form using additives.

(2) Sintered calcine - a process to produce a sintered glass and crystalline phase product, using fluxes and binders.

(3) Metal Matrices - a process to produce a high thermal conductivity monolithic form of calcine, vitreous beads, or pellets cast in molten metal or embedded in sintered metal.

(4) Glass/Ceramic - a process producing a glass subjected to controlled crystallization, thus ending up with a fine grained crystalline body product.

(5) Coated Pellets - a process to take calcine powders and coat them with a durable material such as Al_2O_3 or glass and sinter them to produce stable pellets.

(6) Aqueous silicate - a process to convert wastes to solid, relatively insoluble, aluminosilicate minerals. Powdered clays such as kaolin or bentonite are mixed with, and allowed to react with, aqueous solutious or slurries of caustic wastes at temperatures ranging from 30-100 C.

5.0 Cited References

1. Statement by President J. E. Carter on "Nuclear Power Policy," April 7, 1977, White House, Washington, D. C. (see also Nucleonics Week, Vol. 18, No. 15, April 14, 1977).

2. Report of Task Force for Review of Nuclear Waste Management - Draft, Report DOE/ER-0004/D, U. S. Department of Energy, Washington, D. C., February 1978.

3. Alternatives for Long-Term Management of Defense High-Level Radioactive Waste, Idaho Chemical Processing plant, Report ERDA 77-43, U. S. Energy Research and Development Administration, Washington, D. C., September 1977.

4. Final Environmental Impact Statement, Waste Management Operations, Idaho National Engineering Laboratory, Idaho, Report ERDA-1536, U. S. Energy Research and Development Administration, Washington, D. C., September 1977.

5. INEL Waste Management Plan for FY-1978 Idaho National Engineering Laboratory, Report IDO-10051, U. S. Energy Research and Development Administration, Idaho Falls, Idaho, August 1977

6. (a) Alternatives for Long-Term Management of Defense High-Level Radioactive Waste, Savannah River Plant, Report ERDA 77-42, U. S. Energy Research and Development Administration, Washington, D. C., May 1977. (b) Environmental Statement, Waste Management Operations, Savannah River Plant, Aiken, South Carolina, Report ERDA-1537, U. S. Energy Research and Development Administration, Washington, D. C., September 1977

7. Alternatives for Long-Term Management of Defense High-Level Radioactive Waste, Hanford Reservation, Report ERDA 77-44, U. S. Energy Research and Development Administration, Washington, D. C., September 1977.

8. Final Environmental Statement, Waste Management Operations, Hanford Reservation, Richland, Washington, Report ERDA-1538, 2 Volumes, U. S. Energy Research and Development Administration, Washington, D. C., December 1975.

9. Alternative Processes for Managing Existing Commercial High-Level Radioactive Wastes, Report NUREG-0043, U. S. Nuclear Regulatory Commission, Washington, D. C., April 1976.

10. G. J. Dau and R. F. Williams, Status of Commercial Nuclear High-Level Waste Disposal, Report EPRI-NP-44-SR, Electric Power Research Institute, Palo Alto, California, September 1976.

11. Alternatives for Managing Wastes from Reactors and Post Fission Operations in the LWR Fuel Cycle, Volume 2: Alternatives for Waste Treatment, Report No. ERDA-76-43, U. S. Energy Research and Development Administration, Washington, D. C., May 1976.

12. High-Level Radioactive Waste Management Alternatives, Report No. WASH-1297, U. S. Atomic Energy Commission, Washington, D. C., May 1974.

13. W. J. George, "Treating and Disposing of Radioactive Wastes," Chemical Engineering, page 151, December 14, 1959.

14. "Management of Radioactive Wastes," Atomics, page 16, May/June, 1965.

15. B. R. Wheeler, et al., A Comparison of Various Calcination Processes For Processing High-Level Radioactive Wastes, Report IDO-1422, U. S. Atomic Energy Commission, Idaho Falls, Idaho, April 1964.

16. R. E. Tomlinson (Ed), Radioactive Wastes From the Nuclear Fuel Cycle, Symposium Series 154, Volume 72, Americal Institute of Chemical Engineers, New York, N. Y., 1976.

17. Management of Radioactive Wastes from Fuel Reprocessing, Proceedings of a Symposium organized jointly by the OECD Nuclear Energy Agency and the International Atomic Energy Agency, Paris, November 27-December 1, 1972.

18. A. M. Platt and C. R. Cooley, "Waste Solidifiation Prototype Program," Atomics, page 22, July/August 1965.

19. K. J. Schnieder, "Solidification of Radioactive Waste," Chemical Engineering Progress, Vol. 66, No. 2, page 35, February 1970.

20. B. R. Dickey, et al., "High-Level Waste Solidification: Applicability of Fluidization-Bed Calcination to Commercial Wastes," Nuclear Technology, Vol, 24, No. 3, p. 371, December 1977.

21. W. F. Holcomb, "Uses of the Fluidization Bed Process," Combustion, Vol 48, No. 10, page 31, April 1977.

22. J. A. Wielang and W. A. Freeby, The Fifth Processing Campaign in the Waste Calcining Facility-FY 1972, USAEC Report No. ICP-1021, Idaho National Engineering Laboratory, Idaho Falls, Idaho, June 1973.

23. W. F. Holcomb and J. A. Wielang, "O_2 for Calcification of Radioactive Wastes," Cryogenics and Industrial Gases, Vol. 8 No. 4, page 24, July/August 1973.

24. L. T. Lakey and B. R. Wheeler, "Solidification of High-Level Radioactive Wastes at the Idaho Chemical Processing Plant," in reference 17, page 731.

25. A. G. Blasewitz et al., "The High-Level Waste Solidification Program:, in reference 17, page 615.

26. F. Kaufmann, et al., "Recent Experience with a Steam-Heated Spray Calcining Unit for High-Level Waste Solidification," in reference 16, page 132.

27. W. Guber et al., "Pilot Plant Experience on High-Level Waste Solidification and Design of the Engineering Prototype VERA," in reference 17, page 489.

28. R. Bonniaud et al., "French Industrial Plant AVM for Continuous Vitrification of High-Level Radioactive Wastes", in reference 16, page 145.

29. J. A. Stone, Evaluation of Concrete as a Matrix for Solidification of Savannah River Plant Waste, USERDA Report DP-1448, Savannah River Laboratory, Aiken, South Carolina, June 1977.

30. M. J. Plodinec, Evaluation of Cs-137 Sorbents for Fixation in Concrete, USERDA Report DP-1444, Savannah River Laboratory, Aiken, South Carolina, February 1977.

31. N. E. Bibler, Radiolytic Gas Production From Concrete Containing Savannah River Plant Waste, USDOE Report DP-1464, Savannah River Laboratory, Aiken, South Carolina, January 1978.

32. J. A. Stone and P. D. d'Entremont, Measurement and Control of Cement Set Times in Waste Solidification, USERDA Report DP-1404, Savannah River Laboratory, Aiken, South Carolina, September 1976.

33. J. A. Stone, "Evaluation of Concrete as a Matrix for Solidification of Savannah River Plant Waste," presented at the 80th Annual meeting of the American Ceramic Society, Detroit, Michigan, May 6-11, 1978.

34. Ceramic and Glass Radioactive Waste Forms, Workshop Summary, Report No. CONF-770102, U. S. Energy Research and Development Administration, Washington, D. C., January 4-5, 1977.

35. R. B. Keely et al., "Technology Status of Spray Calcination/Vitrification of High-Level Liquid Waste for Full-Scale Application," Presented at the 70th Annual Meeting of the American Institute of Chemical Engineers, New York, November 13-17, 1977.

36. C. C. Chapman et al., "Experience with Waste Vitrification Systems at Battelle-Northwest," in reference 16, page 151.

37. D. E. Larson and W. F. Bonner, High-Level Waste Vitrification by Spray Calcination/In-Can Melting, USAEC Report BNWL-2092, Battelle Pacific Northwest Laboratories, Richland, Washington, November 1976.

38. J. E. Mendel, "Glass as a Waste Form: Overview and Processing Considerations," in reference 29, page 29.

39. R. N. Rickles, "The Encapsulation of High-Level Nuclear Wastes," Atomics, page 50, March/April 1964.

6.0 Additional References

1. J. E. Mendel, et al, Annual Report on the Characteristics of High-Level Waste Glasses, USERDA Report BNWL-2252, Battelle Pacific Northwest Laboratories, Richland, Washington, June 1977.

2. W. J. Gray, Volatility of a Zinc Borosilicate Glass Containing Simulated High-Level Radioactive Waste, USERDA Report BNWL-2111, Battelle Pacific Northwest Laboratories, Richland, Washington, October 1976.

3. H. T. Blair, Vitrification of Nuclear Waste Calcines by In-Can Melting, USERDA Report BNWL-2061, Battelle Pacific Northwest Laboratories, Richland, Washington, May 1976.

4. J. E. Mendel, "High-Level Waste Glass," Nuclear Technology, Vol. 32, No. 1, page 72, January 1977.

5. J. A. Kelley, Evaluation of Glass as a Matrix for Solidification of Savannah River Plant Waste, USERDA Report DP-1397, Savannah River Laboratory, Aiken, South Carolina, October 1975.

6. Management of Radioactive Wastes From the Nuclear Fuel Cycle, Proceedings of a Symposium jointly organized by the IAEA and NEA (OECD), Vienna, Austria, March 22-26, 1976.

7. C. C. Chapman and J. L. Buelt, "Vitrification of High-Level Waste in a Joule Heated Ceramic Melter," Presented at the 70th Annual Meeting of the American Institute of Chemical Engineers, New York, November 13-17, 1977.

8. J. R. La Riviere and E. L. Moore, Preliminary Canister Size Criteria for Commercial Solidified High-Level Waste, USERDA Report ARH-ST-151, Atlantic Richfield Hanford Company, Richland, Washington, April 1977.

9. R. P. Turcotte, Radiation Effects in Solidified High-Level Wastes, Part 2-Helium Behavior, USERDA Report BNWL-2051, Battelle Pacific Northwest Laboratories, Richland, Washington, May 1976.

10. G. J. McCarthy, "High-Level Waste Ceramics: Materials Considerations, Process Simulation, and Product Characterization," Nuclear Technology, Vol, 32, No. 1, p 92, January 1977.

11. W. K. Eister, "Materials Considerations in Radioactive Waste Storage," Nuclear Technology, Vol 32, No. 1, page 6, January 1977.

12. S. C. Slate and W. A. Ross, "High-Level Radioactive Waste Glass and Storage Canister Design," Presented at the 70th Annual Meeting of the American Institute of Chemical Engineers, New York, November 13-17, 1977.

13. R. E. Schindler, et al, Development of a Fluidized-Bed Calciner and Post-Treatment Processes for Solidification of Commercial Fuel-Reprocessing Liquid Wastes, USDOE Report ICP-1136, Idaho National Engineering Laboratory, Idaho Falls, Idaho, December 1977.

14. J. A. Wielang, et al, The Fourth Processing Campaign in the Waste Calcining Facility FY 1971, USAEC Report ICP-1004, Idaho National Engineering Laboratory, Idaho Falls, Idaho, March 1972.

15. W. A. Freeby, Interim Report: Fluidized-Bed Calcination of Simulated High-Level Commercial Wastes, USERDA Report ICP-1075, Idaho National Engineering Laboratory, Idaho Falls, Idaho, June 1975.

16. W. J. Bjorklund, Fluidized Bed Calcination Experience with Simulated Commercial High-Level Nuclear Waste, USERDA Report BNWL-2138, Battelle Pacific Northwest Laboratories, Richland, Washington, November 1976.

17. G. L. Tingey and W. D. Felix, Radiolytic Gas Generation from Calcined Nuclear Waste, USERDA Report BNWL-2381, Battelle Pacific Northwest Laboratories, Richland, Washington, August 1977.

18. J. R. Berreth, "Stabilization and Storage of Solidified High-Level Radioactive Wastes," Nuclear Technology, Vol. 32, page 16, January 1977.

19. Current Activities in DOE's Commercial Waste Management Program, Report DOE/ET-0042, U. S. Department of Energy, Washington, D. C., March 1978.

20. W. P. Bishop and F. J. Miraglia, Jr., Eds., Environmental Survey of the Reprocessing and Waste Management Portions of the LWR Fuel Cycle, Report NUREG-0116 (Supp. 1 to WASH-1248), U. S. Nuclear Regulatory Commission, Washington, D. C., October 1976.

21. The Waste Calcining Facility at the Idaho Chemical Processing Plant, U. S. Atomic Energy Commission, Idaho Falls, Idaho, 1970.

22. W. F. Bonner, et al, Spray Solidification of Nuclear Waste, USERDA Report BNWL-2059, Battelle Pacific Northwest Laboratories, Richland, Washington, August 1976.

23. W. A. Ross, Development of Glass Formulations Containing High-Level Nuclear Waste, Report PNL-2481, Battelle Pacific Northwest Laboratories, Richland, Washington, February 1978.

24. T. H. Smith and W. A. Ross, Impact Testing of Vitneous Simulated High-Level Waste in Canisters Report BNWL-1903, Battelle Pacific Northwest Laboratories, Richland, Washington, May 1975.

25. "A New Glass Container for the Safe Encapsulation of Radioactive Wastes from Nuclear Power Plants Has Been Invented by Three Catholic University Professors," Atomic Energy Clearing House, Vol. 23, No. 43, page 9, Washington, D. C., October 24, 1977.

26. The Management and Storage of Commercial Power Reactor Wastes, Report ERDA-76-162, U. S. Energy Research and Development Administration, Washington, D. C., 1976.

27. G. Malow and W. Lutze, "Sodium diffusion in borosilicate glasses. Part 1. Influence of glass composition," and "Part 2. Ion dipole interactions," Physics and Chemistry of Glasses, Vol. 16, No. 6, December 1975.

28. W. Heimerl, et al, Research on Glasses for Fission Product Fixation, Summary Report January 1968-June 1971, Report No. HMI-B-109, Hahn-Mertner-Institute Berlin GmbH, Federal Republic of Germany, September 1971.

29. D. W. Clelland, et al, "Design of a Plant for the Incorporation of Highly Active Wastes Into Glass," presented at the 60th Annual Meeting of the American Institute of Chemical Engineers, New York, N.Y., November 26-30, 1967.

30. F. Laude, "Pot Vitrification of Concentrated Fission Product Solutious," symposium proceedings on Solidification and Long-Term Storage of Highly Radioactive Wastes, U. S. Atomic Energy Commission, Richland, Washington, 1966.

31. P. R. Savage, "Nuclear Waste disposal: politics closed prospects," Chemical Engineering, Vol. 84, page 72, June 20, 1977.

32. M. D. Alford, Fluidized Bed Calcination of Simulated Purex High-Activity Level Wastes, Interim USAEC Report No. HW-66384 RD, General Electric Hanford Atomic Products Operation, Richland, Washington, September 1960.

33. M. D. Alford and K. J. Schneider, Prototype Waste Calciner, USAEC Report No. HW-62631, General Electric Hanford Atomic Products Operation, Richland, Washington, November 1959.

34. W. A. Ross and J. E. Mendel, "Glass Waste Forms for Radioactive Waste Containment," Presented at the 70th Annual Meeting of the American Institute of Chemical Engineers, New York, November 13-17, 1977.

35. K. M. Lamb, Final Report: Development of a Metal Matrix for Incorporating High-Level Commercial Waste, USDOE Report ICP-1144, Allied Chemical Corporation, Idaho Chemical programs, Idaho Falls, Idaho, March 1978.

36. "Waste Solidification Gains Major Attention," Nucleonics, Vol, 21, No. 2, page 58, February 1963.

37. J. E. Mendel, et al., A Program Plan for Comprehensive Characterization of Solidified High-Level Wastes, USAEC Report BNWL-1940, Battelle Pacific Northwest Laboratories, Richland, Washington, December 1975.

38. A. K. De, et al., "Development of Glass Ceramics for the Incorporation of Fission Products." Ceramic Bulletin, Vol. 55, No. 5, page 500, 1976.

39. G. H. Thompson, Interim Solidification of SRP Waste With Silica, Bentonite, or Phosphoric Acid, USERDA Report DP-1403, Savannah River Laboratory, Aiken, South Carolina, March 1976.

40. C. M. Slansky, Ed., Technical Division Quarterly Progress Report, October-December 1977, USDOE Report ICP-1141, Idaho National Engineering Laboratory, Idaho Falls, Idaho, February 1978.

41. A. M. Platt (Compiler), Nuclear Waste Management Quarterly Progress Report, July through September 1977. USDOE Report PNL-2377-3, Battelle Pacific Northwest Laboratories, Richland, Washington, January 1978.

42. J. L. McElroy, Quarterly Progress Report, REsearch and Development Activities Waste Fixation Program, January through March 1977, USDOE Report PNL-2265-1, Battelle Pacific Northwest Laboratories, Richland, Washington November 1977.

43. Management of Wastes from the LWR Fuel Cycle, Proceedings of the International Symposium, Denver, Colorado, Report CONF-76-0701. U. S. Energy Research and Development Administration, Washington, D. C., July 11-16, 1976.

44. T. English, et al, An Analysis of the Technical Status of High-Level Radioactive Waste and Spent Fuel Management Systems, Report JPL-77-69, Jet Propulsion Laboratory, Pasadena, California, December 1977.

45. "Waste Management: Technological Advances and Attitudes of Safety," Nuclear News, Vol 7, No. 10, p 94, October 1964.

46. Radioactive Waste Management - A Bibliography of Publicly Available Literature Pertaining to the USAEC's Hanford, Washington Production Site, Report TID-3340, U. S. Atomic Energy Commission, Oak Ridge, Tennessee, August 1973.

47. Radioactive Waste Management - A Bibliography of Publicly Available Literature Pertaining To the USAEC's Savannah River Plant, South Carolina, Report TID-3341, U. S. Atomic Energy Commission, Oak Ridge, Tennessee, August 1973.

48. Radioactive Waste Management - A Bibliography of Publicly Available Literature Pertaining to the USAEC's National Reactor Testing Station, Idaho, Report TID-3342, U. S. Atomic Energy Commission, Oak Ridge, Tennessee, August 1973.

49. W. G. Belter and D. W. Pearce, "Radioactive Waste Management," Reactor Technology - Selected Reviews - 1965, Report TID-8541, U. S. Atomic Energy Commission, Oak Ridge, Tennessee, January 1966.

50. C. L. Bendixsen, et al, The Third Processing Campaign in the Waste Calcining Facility, USAEC Report IN-1474, Idaho National Engineering Laboratory, Idaho Falls, Idaho, May 1971.

51. Siting of Fuel Reprocessing Plants and Waste Management Facilities, USAEC Report ORNL-4451, Oak Ridge National Laboratory, Oak Ridge, Tennessee, July 1970.

52. G. E. Lohse, et al, "Calcination of Zirconium Fluoride Wastes in Pilot Scale and Plant Scale Fluidised Beds," International Chemical Engineering Symposium Series No. 30 London, 1968.

53. K. J. Schneider, Editor, Waste Solidification Program,

Volume 1	Report BNWL-1073	August 1969
Volume 2	Report BNWL-968	February 1969
Volume 3	Report BNWL-832	December 1968
Volume 4	Report BNWL-814	December 1968
Volume 5	Report BNWL-1185	January 1970
Volume 6	Report BNWL-1391	August 1970
Volume 7	Report BNWL-1541	January 1971
Volume 8	Report BNWL-1583	June 1971
Volume 9	Report BNWL-1628	January 1972

Battelle Pacific Northwest Laboratories, Richland, Washington.

54. J.N.C. van Geel, et al, "Conditioning High-Level Radioactive Waste," Chemical Engineering Progress, Vol 72, No. 3, P 49, March 1976.

55. R. E. Blanco, et al, "Solving the Waste-Disposal Problem," Nucleonics, Vol 25, No. 2, p 58, February 1967.

56. "Calcining Techniques to Ease Nuclear-Waste Woes," Chemical Engineering, p 26, April 1, 1963.

57. "A Glass Trap for Nuclear Wastes," Chemical Engineering, p 84, July 22, 1963.

58. E. D. Cooper et al, "Pilot - and Plant-Scale Fluidized Bed Calciners", Chemical Engineering Progress, Vol 61, No. 7, p 89, July 1965.

59. "AEC Well Along on Waste Disposal," Nucleonics, p 78, February 1964.

60. B. L. Cohen, "The Disposal of Radioactive Wastes from Fission Reactors", Scientific American, Vol 236, No. 6, p 21, June 1977.

61. K. M. Harmon, Summary of National and International Radioactive Waste Management Programs, USDOE Report PNL-2598, Battelle Pacific Northwest Laboratories, Richland, Washington, March 1978.

62. J. R. Berreth, et al., Status Report: Development and Evaluation of Alternative Treatment Methods for Commercial and ICPP High-Level Solidified Wastes, USERDA Report ICP-1089, Idaho National Engineering Laboratory, Idaho Falls, Idaho, May 1976.

63. J. A. Powell, (Ed), Bibliography of PNL Publications in Managment of Radioactive Wastes, USERDA Report BNWL-2201, Battelle Pacific Northwest Laboratories, Richland Washington, July 1976.

64. W. A. Ross, et al, Annual Report on the Characterization of high-Level Waste Glasses USDOE Report PNL-2625, Pacific Northwest Laboratory, Richland, Washington, June 1978.

65. R. O. Lokken, A Review of Radioactive Waste Immobilization in Concrete, USDOE Report PNL-2654, Battelle Pacific Northwest Laboratory, Richland, Washington, June 1978

IV. LOW-LEVEL RADIOACTIVE WASTE SOLIDIFICATION TECHNIQUES

Due to the increasing amount of waste being generated both in volume and activity by the nuclear industry, considerable interest has been shown during the last several years concerning various methods and systems for the solidification of liquid and solid low-level radioactive wastes from nuclear power plants; and, to a varying degree, from other fuel cycle facilities. The present U.S. method for the disposal of low-level radioactive wastes is by burial in shallow trenches dug in the earth's surface.

There are many available varieties of solidification materials and techniques. Solidification agents include portland cement, concrete, plaster of paris, asphalt (or bitumen), polymers, and a blend of absorbent material and cement or plaster (1-7). Presently, most of the solidification systems used in the United States utilize either cement or an organic polymer, such as urea formaldehyde, as the basic solidification matrix material.

Table I lists the various solidification agents and the process vendor or developer.

Table I

SOLIDIFICATION AGENTS AND VENDORS

Vendor or Developer	Solidification Agent
Aerojet Energy Conversion Co. Sacramento, California	Cement Urea Formaldehyde Bitumen
ANEFCO, Inc. White Plains, New York	Urea Formaldehyde
ATCOR, Inc. (a) Peekskill, New York	Cement
Brookhaven National Laboratory (b) Upton, Long Island, New York	Cement with organic polymers
Chem-Nuclear Systems, Inc. Bellevue, Washington	Cement Urea Formaldehyde
Delaware Custom Materials Cleveland, Ohio	Cement with shale or silcates
Dow Chemical Company Midland, Michigan	Organic Polymer
Energy, Inc. Idaho Falls, Idaho Newport News Industrial Corp. Newport News, Virginia	Cement Urea Formaldehyde and Bitumen
General Electric Company San Jose, California	Cement
Hittman Nuclear & Development Corp Columbia, Maryland	Cement Urea Formaldehyde
Los Alamos Scientific Laboratory (b) Los Alamos, New Mexico	Cement

Oak Ridge National Laboratory (b) Oak Ridge, Tennessee	Cement with additives
Protective Packaging, Inc. Jeffersontown, Kentucky	Urea Formaldehyde
Stock Equipment Company Cleveland, Ohio	Cement Urea Formaldehyde
Todd Research and Technical Div. Galveston, Texas	Organic polymer
United Nuclear Industries Richland, Washington	Cement with sodium silicate Urea Formaldehyde
United Technologies Sunnyvale, California	Organic polymer
Werner and Pfleiderer Corp. Waldwick, New Jersey	Bitumen
Westinghouse Electric Corp. Pittsburg, Pennsylvania	Cement with vermiculite

a/ A division of Chem-Nuclear Systems, Inc.,
See Nucleonics Week, August 31, 1978

b/ Non-commercial applications

Wastes from the nuclear fuel cycle and particularly from power plants can be solid, liquid or gaseous with varying chemical, physical, and radiological characteristics with many systems processing special liquids which produce wet solids such as evaporator concentrates, resins, etc.

The radioactive liquid wastes from the power reactors, such as, primary system blowdown, equipment drains, resin sluicing water, evaporator condensates, decontamination solutions, demineralizer regenerative solutions, laundry and laboratory wastes, evaporator concentrates, spent ion exchange resins, filter precoat and cake materials (powdex and solka-floc), cartidge filter units, and diatomaceous earth are suitable for immobilization. These wastes contain the bulk of the volume and radioactivity of the solidified wastes sent to the commercial low-level burial facilities.

The processing of these wastes can be broken down into five steps: (a) waste collection; (b) pretreatment; (c) the solidification process; (d) mixing and packaging; and (e) final handling (Figure 7) (3,8). Collection usually takes place in sumps or tanks; the contents are then processed on batch or semi-continuous basis. The solids pretreatment operation consists of reducing the volume of the wet solids by using an evaporator or other volume reduction device. The solidification and mixing steps involve the use of an agent, such as cement or an organic polymer with additives or a catalyst, to produce an immobilized, monolithic, inert matrix. The container handling operations include inspection to ascertain that solidification took place, capping the container and adding appropriate shielding, decontamination, marking and labeling, container testing, and storage awaiting shipment to a burial facility.

1.0 Solidification Of Low-Level Radioactive Wastes In Bitumen

The use of bitumen to solidify low-level radioactive wastes has been successfully applied on an industrial scale for many years in different countries (1-5, 9-19). Bitumen or asphalt, a mixture of high-molecular weight hydrocarbons, is a by-product residue from the petroleum refining processes. Various grades of bitumen are commercially available with a wide range of physical properties. Bitumen processes generally operate in the range of 150 to 230° C, at which temperature water originally present in the waste potentially can be volatilized.

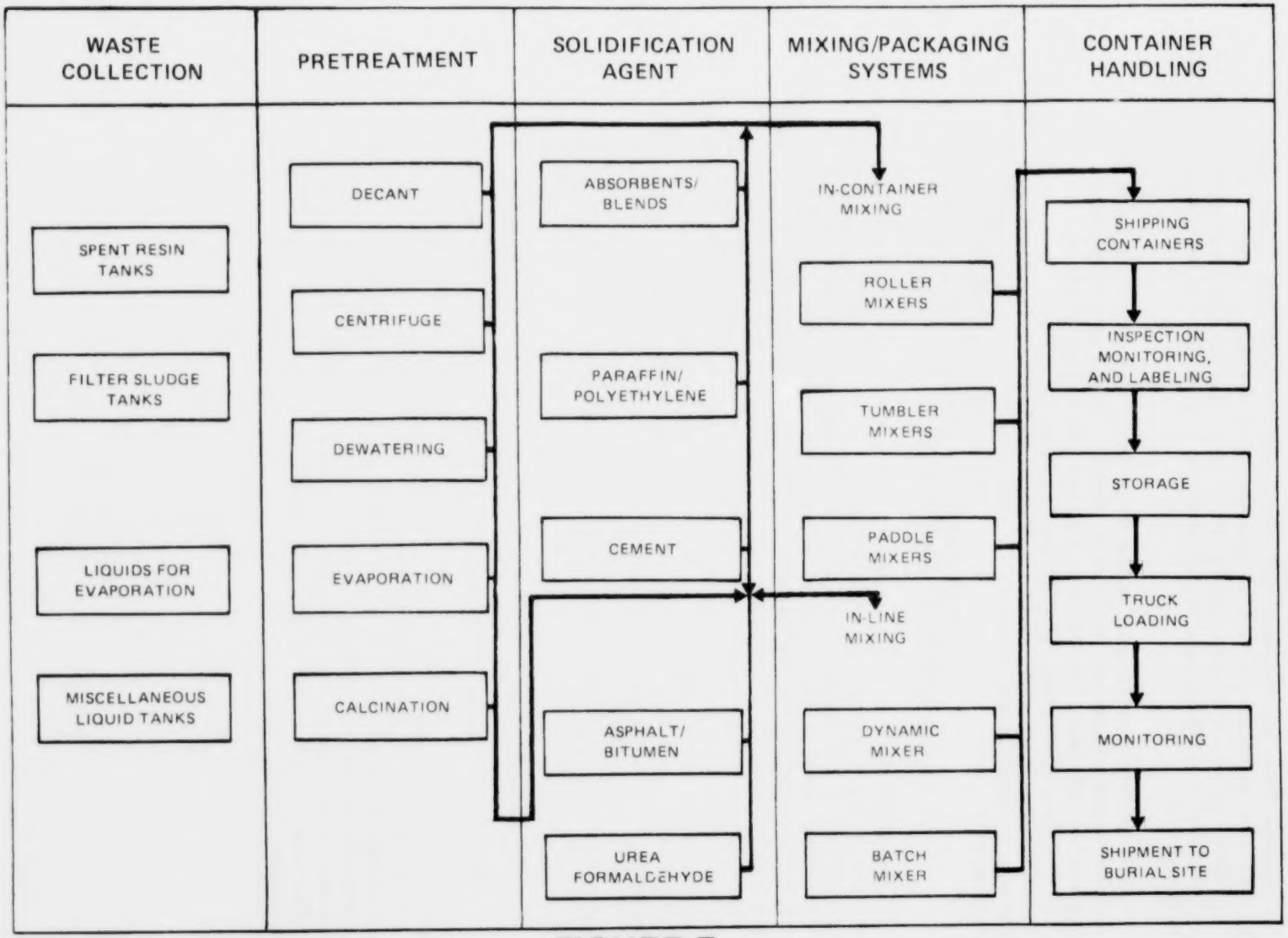

FIGURE 7

LOW-LEVEL WASTE PROCESSING STEPS

Basically, all of the bitumen processes consist of mixing the waste solution, slurry, or solids with commercial emulsified asphalt or molten base-asphalt and raising the temperature to evaporate the fluid. The contaminated solids remain intimately dispersed in the asphalt and the product flows out of an evaporator into a receptacle. The process has been successfully demonstrated in both continuous and batch mixing operations. Four different processes have been developed for the bitumen-waste incorporation process: (a) stirred evaporation; (b) film evaporation; (c) the emulsified bitumen process; and (d) the screw extrusion. Several versions of the bitumen processes for incorporating radioactive wastes for disposal have been utilized on an industrial scale in Europe and on a laboratory development basis in the U. S.

1.1 Stirred-Evaporator Batch Process

In this process the bitumen is preheated and charged to an evaporator with waste being added in with the bitumen. Electric heat is provided by immersion heaters and through the vessel wall. An agitator with adjustable blades is used for mixing and homogenizing the waste-bitumen charge. The charge is kept in the evaporator for several hours, prior to discharging to a disposal container. Figure 8 illustrates this process. The first plant scale bitumen process, using the stirred evaporator method was started in 1964 with an evaporation rate of 100 liters per hour at Mol, Belgium (1,4). The initial operation was directed at the bitumen incorporation of radioactive chemical sludges. Subsequently, concentrated solutions, incinerator ash, vermiculite, ion-exchange material and sand have been processed using a slow-mixing device, particle separation, and resin incineration. Certain oxidizing salts, particularly nitrates, produce an undesirable hardness in the final product; however, this difficulty is overcome by use of reducing agents or different bitumen mixes. Boric acid solutions can be incorporated into bitumen if the solutions are first neutralized to prevent acid volatilization and water leaching in the final product.

1.2 Emulsified Bitumen Process

In this process, as shown in Figure 9, the waste material is concentrated by filtration on a rotary filter then fed to a mixing station where the bitumen and surface-active agents are added. Here the mixing and coating of the solid waste particles takes place with separation of the water. Upon removal of the water, the mix goes to a drier. The remaining water is reduced and the product discharged to drums.

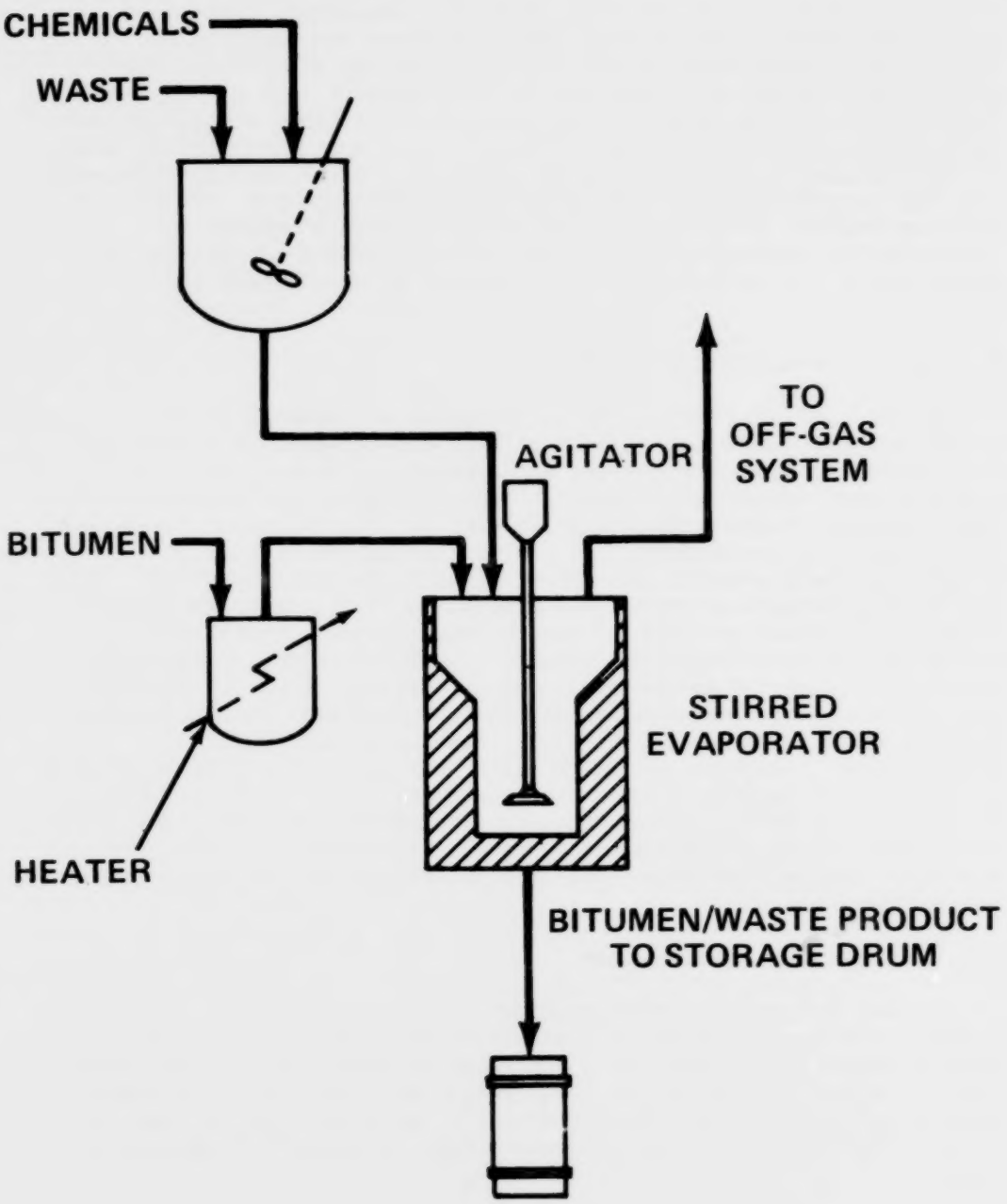

FIGURE 8

STIRRED–EVAPORATOR BATCH PROCESS

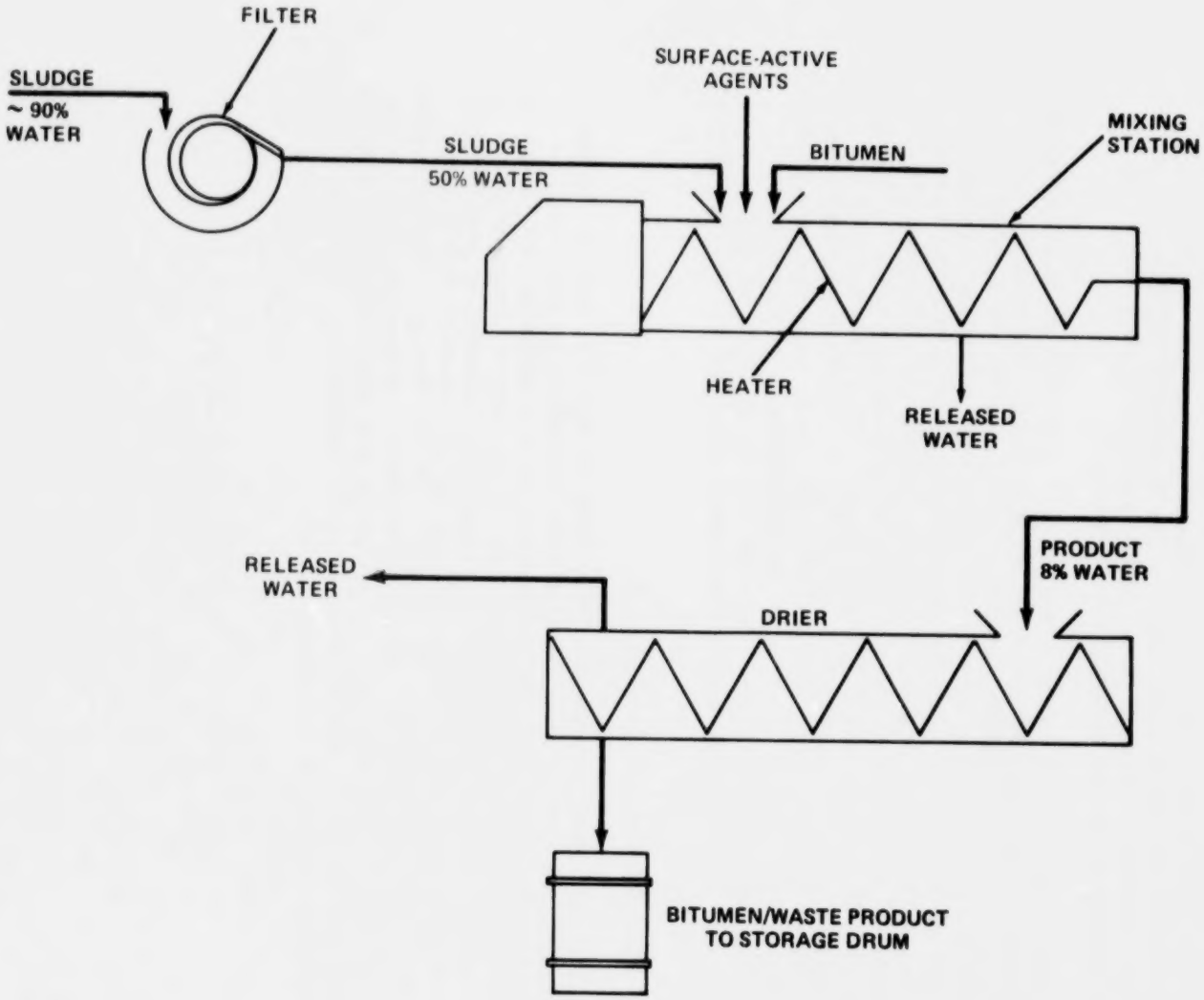

FIGURE 9

EMULSIFIED BITUMEN PROCESS

This process was pioneered at the Marcoule Centre, France for radioactive sludges and requires a much higher bitumen content. Various surface-active agents (a surface-active agent is a soluble compound that reduces the surface tension of liquids, or reduces interfacial tension between two liquids or between a liquid and a solid), variable reaction times, and different bitumen varieties are used depending on the sludge to be treated.

Experimental work on this process was also done at the Department of Energy's Oak Ridge National Laboratory (ORNL), Tennessee (4, 10). In this process the initial mixing of liquid waste and bitumen could be effected readily at any convenient temperature below the boiling point of the waste solution with the water and/or volatiles being removed by heating. The liquid wastes of special interest to ORNL were the evaporator concentrates and solutions of sodium metaborate, nitrate and nitrite. No significant difficulities were experienced in the incorporation of 60 weight per cent (w/o) of solids in bitumen from evaporator concentrates; however, the best final products were produced at solids contents of 45-50 w/o. Boron compounds required higher temperatures, neutralization, and low sodium to boron ratios, especially with tetraborates which harden the bitumen, making stirring impossible.

1.3 Film Evaporation Process

This process has been developed both in the U.S. and in foreign countries, with an industrial-size unit operating in France. It consists of a vertical tube evaporator with a steam-heated jacket and internal rotor that forms a thin film of the waste-bitumen mixture on the heated surface. The waste plus preheated bitumen is continuously fed to the unit. The water is evaporated and removed from the top, while the bitumen-waste is discharged from the bottom. The residence time in the evaporator is only a few minutes. Figure 10 illustrates this process. ORNL has also investigated the film evaporator process, along with Marcoule, for incorporation of industrial, urban and radioactive wastes in asphalt (10, 17).

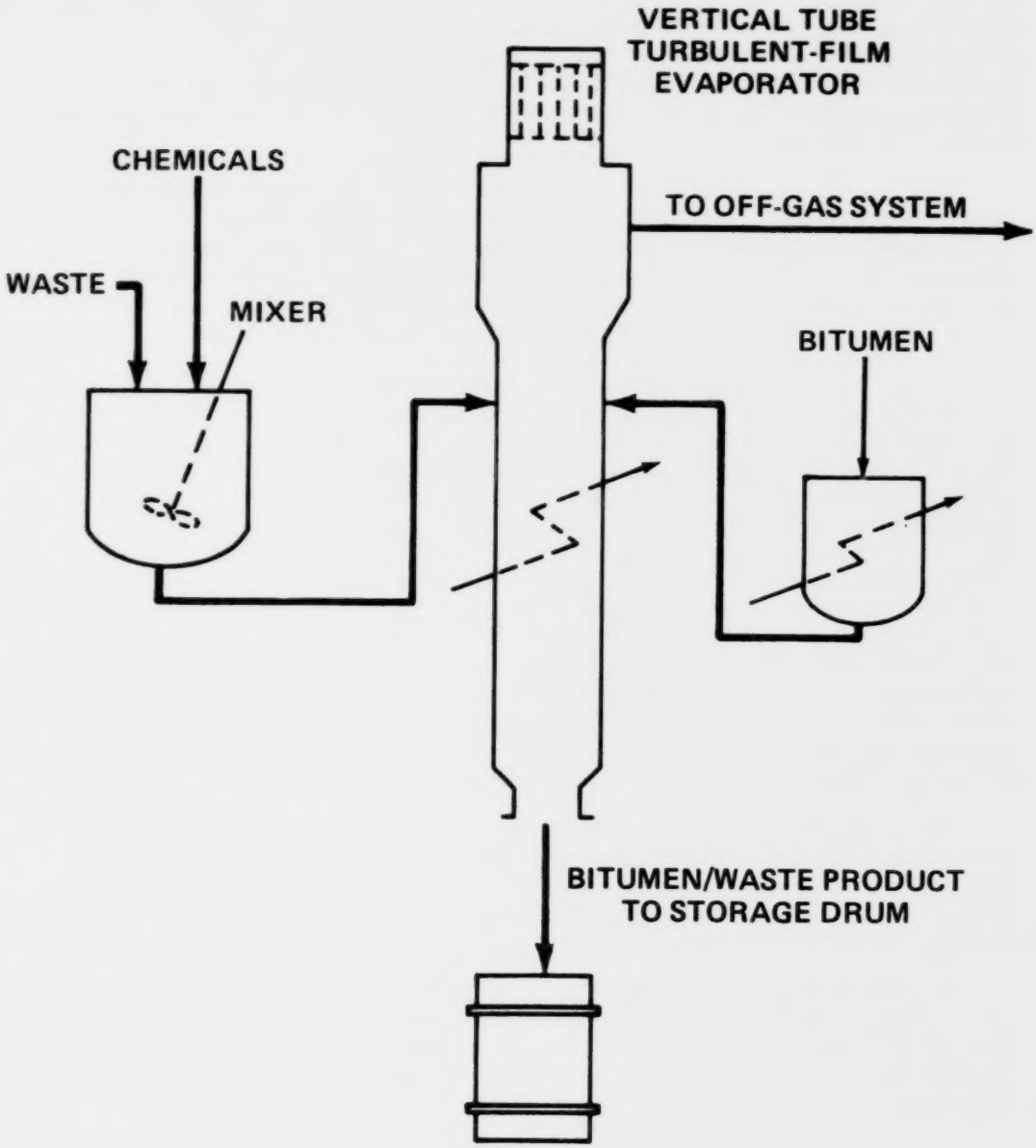

FIGURE 10

TURBULENT-FILM EVAPORATION PROCESS

1.4 Screw Extrusion Process

The screw extruder evaporator concept, as shown in Figure 11, was developed at the Eurochemic Plant at Mol, Begium and at the Karlsruhe Nuclear Research Center in West Germany with good success at both facilities (11, 16). Industrial bituminization plants for solidification of evaporator concentrates, utilizing the screw-extruder-evaporator, have been sucessfully operated at an evaporation capacity of 140 kg of water per hour with the final product containing 50% salts (14-16).

This process consists of two or four screw extruders operating in a steel barrel that is divided into a number of stages heated individually by steam. Waste and preheated bitumen are charged directly into the barrel. While the screws mix the waste-bitumen mixture and transport it forward, the water is evaporated and removed. The product is discharged into drums and allowed to cool before capping.

Werner & Pfleiderer Corporation (WPC) offers a waste solidification system that includes a volume reduction system (3, 11, 14-16, 18, 19). With asphalt as the solidification agent, WPC indicates the end product is stable, particularly against leaching. Although it has only been recently introduced in North and South America, and Asia, it has been operating routinely in Europe since 1965. The WPC radwaste solidification process yields a liquid-free (less than 0.5%) solid using a continuous, fully automatic process with a multi-screw compounding extruder. The extruder-evaporator simultaneously provides homogenous mixing (including reagent additives), liquid evaporation and solidification in one machine. The extruder evaporator normally discharges the asphalt/salts mix into standard DOT 208 liter drums at a rate from 1 to 114 liters per hour, depending on the size and speed of the extruder and the concentration of the feed stream. The entire process, complete with interlocks, can be controlled remotely.

The extruder-evaporator system can also evaporate the excess water away from powdered resin sludge and bead resins in addition to evaporator concentrates. This system can also be used, to some extent, for volume reduction and solidification of diatomaceous earth filter sludge, depending on the quantity.

The WPC extruders, originally developed for the plastics industry, are designed and built to operate a full year without maintenance. The operating record established in Eurpoe bears this out and is quite impressive (over 134,00 hours operation at Marcoule without mechanical failure).

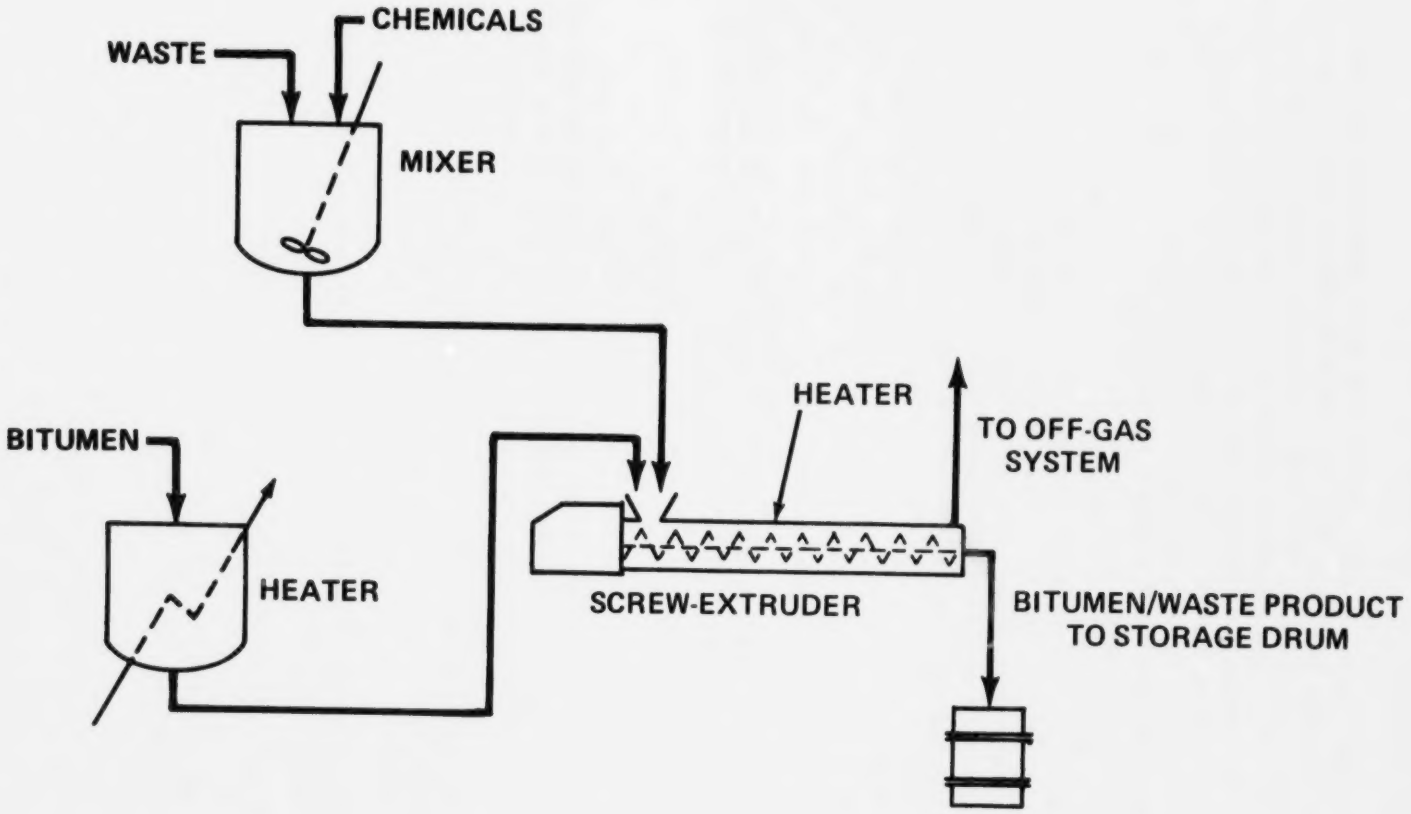

FIGURE 11

SCREW EXTRUDER EVAPORATION PROCESS

1.5 Other Research

Additional research and operational work involving radioactive wastes in bitumen has been done by other countries and nuclear facilities; specifically, the USSR, Bulgaria, Japan, Hungary, and Austria (4, 13). The Hungarian Mineral, Oil and Natural Gas Research Institute developed a technique for the incorporation of nitrate-containing wastes. The technique utilizes an inert gas introduced during the process which eliminates the danger of fires and explosions; the gas also ensures adequate mixing which results in a more satisfactory dispersion of the salts in the bitumen. The Soviet Union has a great deal of interest in the solidification of radioactive concentrates by incorporation in bitumens; they have had a pilot plant in operation since 1969 to evaluate the conditions and feasibility of incorporatiang wastes enriched in sodium nitrate. Also, both the British and Russians have developed methods for immobilizing radioactive wastes attached to natural and synthetic sorbents such as vermiculites, zeolites, clinoptilolite, and ion exchange resins, into a bitumen matrix.

1.6 Advantages and Disadvantages

The main reported advantages and disadvantages of using bitumen for the insolubilization of radioactive wastes are as follows (3, 4, 9, 13, 18):

Advantages:

(a) The leach rate of the final product can be expected to be between 100 and 1000 times lower than cement mixes with a similar concentration of radioactivity.

(b) There are a large number of types of bitumen with a wide variety of properties; thus it is usually possible to obtain a suitable material for any waste.

(c) Bitumen has good coating characteristics and good adhesion to incorporated material.

(d) The solubility of bitumen in water is negligible.

(e) Bitumen possesses a degree of plasticity and elasticity which are of benefit during the incorporation process.

(f) The volume reduction of bitumen compared to cement can be much greater than 5 to 1, depending upon the percentage of radioactive salts or percentage of water in the feed.

(g) Bitumen coats the inside surfaces of the drum, providing resistence to internal corrosion.

(h) Attack by microorganisms on bitumen and bitumen compounds has been regarded as generally insignificant.

Disadvantages

(a) There is always an inherent risk in working with organic material at elevated temperatures; however, there is little evidence that incorporation of the waste material into bitumen increases the risk of fire or explosion. (Tests indicate that the ignition temperature for normal bitumen/salt ratios is above 350°C.)

(b) There is some evidence that the presence of nitrates or nitrites and other oxidizing agents increases the risk of explosions or combustibility of bitumen; however, in laboratory tests neither the high thermal loads nor detonating charges led to explosion-like reactions.

(c) It is obvious that no substance should be added to bitumen which decomposes at the working temperature. Difficulties may be experienced with certain plastics and compounds such as sodium citrate.

(d) Heating of bitumen mixes can result in the releases of oils, fumes and mercaptans.

(e) It is necessary to work at high temperatures to obtain efficient mixing.

(f) To obtain the best final product it is necessary to remove as much as possible of the water present in the waste. Leachability increases significantly wth increasing amounts of retained water.

(g) Strict temperature control is required in the bituminization process.

(h) Mixing tetraborates and iron and aluminium salts with bitumen causes hardening to an extent which can interfere with discharge of the final product from the equipment.

(i) Irradiation of bitumen modifies the chemical and physical properties. In some cases the effect is negligible and in other considerable. Irradiation up to an integrated dose of 10^{10} rad by an external source or up to 10^{8} rad due to the incorporation of radionuclides can be accepted based on the type of bitumen chosen.

(j) Phase separation in bitumen mixes is likely to occur more readily than in cement-waste products, particularly during accidental fires.

(k) Experiments at Marcoule and Karlsruhe have shown that swelling of certain bitumen products can occur in water.

2.0 Solidification Of Low-Level Radioactive Wastes In Cement

The cement solidification process, with and without additives, for radioactive waste treatment has been in common practice on an industrial scale at nuclear facilities for many years in different countries (1-4, 8, 9, 12, 19, 20).

Cement has been described as an adhesive substance, lime being the principal constituent, capable of uniting fragments of solid matter into a compact monolithic structure. The most commonly used cement for the incorporation of radioactive wastes is the "Portland" variety. It is obtained by intimately mixing silica-, alumina- and ferric oxide-bearing materials to the lime and burning these materials to a very hard brick and grinding the resulting brick. There are various types of portland cement depending on the fineness of the grinding and on the addition of certain additives or the amount of various constituents (22-24). Portland cement Type I has been most commonly used, but other Types, such as Type V which is resistant to sulfate salt deterioration, have also been employed.

The cement solidification processes basically consist of mixing the cement with waste solution, slurry, or solids either within the receptacle or just prior to being placed into the receptacle. The actual kinetic process leading to the curing of cement is not well

known. However, the mechanism for the setting of cement, according to the literature, has been postulated to include a reaction between water and cement which forms solid particles and causes crystallization of the calcium hydroaluminate, hydroferrite, and hydrosilicate with the crystals giving the strength to the hardened cement. If the cement product is to be in satisfactory condition for transportation or burial it must have adequate compressive strength. It is common practice to neutralize the acid wastes before the cementation process and to control the salt content. Poorly cured cement will crack and spall, causing more surface area to be exposed to leaching conditions (22, 23).

The mixing of the cement with the various radwaste forms, i.e., sludges, resin beads, etc., affects the properties of the product. The strength of the cement will be a function of the total salt content in the sludges,resins, etc., where there is a narrow range in the acceptable values for the ratio of basic and acidic oxides in the final product. In this regard, waste to cement ratios recommended for proper curing vary significantly among both foreign and U.S. users as shown in Table II. USSR studies have shown that in order to produce cement of acceptable structural strength the concentration of sodium nitrate salt should perferably not exceed 130 g per kg of cement (4).

The presence of water, nitrates, sulfates, borates, and other unstable (in a radiation environment) compounds in the cement could give rise to gaseous radiolytic products (12). Gases also could result from volatilization of compounds by elevated temperature in the cement-waste mixture causing voids to form within the crystalline structure.

Some of the U.S. electric utility companies employ a combination of vermiculite and cement to solidify their radwaste. The expanded vermiculite is porous, permitting the infiltration of dry cement into the vermiculite structure. This would act like a sponge absorbing the liquid and giving a better final product than when cement is used alone. Two cement solidification vendors, United Nuclear Industries and Delaware Custom Materials, have developed a process which use sodium silicate as an additive with Portland cement (3, 25, 26). However, the addition of sodium silicate to cement-waste mixtures increases the volume of waste per volume of solid formed.

In the non-commercial area of using cement for solidification, several of the Department of Energy Laboratories use a variety of techniques. Oak Ridge National Laboratory blends their intermediate-level radioactive wastes with a dry mixture of cementitious materials (Portland Cement-Type 1 and fly ash) and clays. Then using a hydrofracturing technique, the resulting grout is pumped into an isolated shale formation and allowed to set. The clays are used as an additive to retard the movement of radioactive nuclides. The dry solids mix is usually blended with the liquid waste solution at a solids to liquid ratio of between 0.7 and 1 Kg per liter. The volume ratio of grout to liquid waste is approximately 1.4 (27-30).

Brookhaven National Laboratory found that the leachability properties of cement could be improved by developing a polymer-impregnated concrete (PIC) matrix (31-34). PIC composites containing tritiated aqueous waste, solid calcine, incinerator ash, aqueous and solid sodium nitrates, reactor wastes, acidic and neutralized fuel reprocessing wastes, and ion-exchange and sludge materials have been produced. The PIC process utilizes a soak impregnation technique to fill the pores with a styrene monomer, which is then polymerized in-situ by heating to 50-70° C. In addition, the mechanical properties of these cements are significantly improved as a result of the styrene impregnation. PIC also possesses improved radiation stability and resistance to chemical attack.

The Los Alamos Scientific Laboratory mixes transuranic sludge materials and non-transuranic neutralized liquid wastes with cement for storage and burial operations (4, 35, 36).

Table II

CEMENTATION PRACTICES AT VARIOUS ESTABLISHMENTS

Establishment	Nature of Waste	Composition of Mixture
France	(a) Evaporator concentrate (400 g/liter)	250 liters sludge 300 kg cement 40 kg vermiculite
	(b) sludge	83 kg sludge 55 kg cement 10 liters water
F. R. Germany	Evaporator concentrate	100 to 110 liters waste 150 to 200 kg cement
USA	(a) Evaporator concentrate (20% solids)	Vermiculite (2.7 M^3) and Portland cement (0.68 m^3)
	(b) Neutralized Concentrate	(i) 75 liters of concentrate, 128 kg cement, 4 kg vermiculite
		(ii) 20 to 35 liters concentrate, 60 to 65 kg of cement
	(c) Evaporator Concentrates	1 to 2 ratio waste to cement
	(d) Evaporator Concentrates	2 to 3 ratio waste to cement with a sodium silicate additive
	(e) Ion Exchange Resins	1 to 1 ratio resin slurry to cement
Czechoslovakia	(a) Sludge with solids content of 20 to 25%	35 liters of sludge 110 kg of cement

	(b) Evaporator concentrates neutralized to pH 6 to 8 (200 g/liter)	10 kg of sludge 5 kg of evaporator concentrate 22 kg of cement
USSR	Evaporator concentrates (Max. 150 g/liter)	130 g of salt of the sodium nitrate-type per kg of cement

As shown in Figure 12, the cement and radwaste could be mixed either within the shipping container or prior to loading the shipping container. Basically there are two mixing techniques used today: in-drum mixers and external mixers (1-7).

2.1 In-Drum Mixer Process

This technique is probably the simplest version. There are two methods: (a) one is to add weights to a drum along with the cement and waste slurry, then the drum is capped and transferred to a tumbling or rolling station where the contents are throughly mixed; (b) another way is to have an external agitator lowered into the drum to blend the waste and cement, either after or during filling. The mixing blade may be a disposable type which is left in the container or may be a removable type. Figure 13 illustrates this process.

Another system that can be characterized as an in-drum mixing process has been developed by Stock Equipment Company (S-E-Co). Since transport of fresh cement has historically presented difficulties due to the premature hardening and resultant incomplete curing of the waste, S-E-Co. has developed a process to overcome this difficulty by mixing in the final storage drum at the rate of 50 to 200 kg of waste per hour. S-E-Co. has concluded that the quantity of cement and/or additive in each 208 liter drum averages about 91 kg and the amount of radwaste averages about 106 liters. For this system, cap removal, filling, cap replacement, and mixing is an automatic operation.

The S-E-Co. Solid Radwaste System is designed and manufactured as a completely integrated system utilizing components which are designed specifically for the service expected rather than attemping to modify standard equipment. The S-E-Co. System is furnished complete for placement into a building and interfacing wih liquid system piping, utilities, etc. The S-E-Co. design packages the solidified radwaste into standard DOT, 208 liter durms. The S-E-Co. waste system consists of: a cement storage hopper; a storage tank to hold liquid wastes that contain concentrated solutions of dissolved solids; a decant tank for filter media, resins, and/or the solid waste slurry from the storage tanks; a drum processing unit which is fully automatic for uncapping the drums, filling the drums with cement, filling from the decant tank, reinsertion of the cap, and for the mixing/tumbling operation (2, 3, 37).

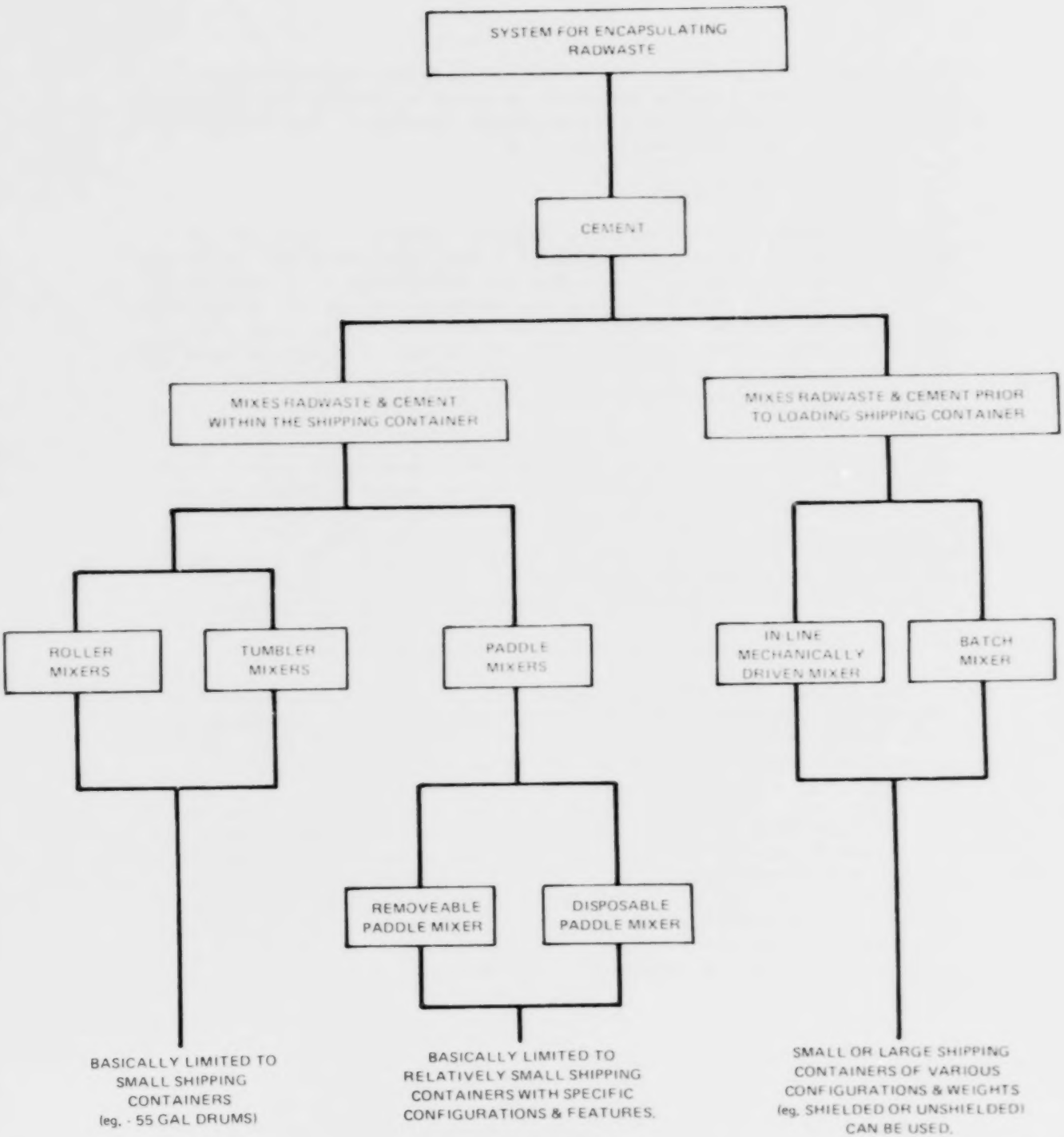
SYSTEM FOR ENCAPSULATING RADWASTE
CEMENT
MIXES RADWASTE & CEMENT WITHIN THE SHIPPING CONTAINER
MIXES RADWASTE & CEMENT PRIOR TO LOADING SHIPPING CONTAINER
ROLLER MIXERS
TUMBLER MIXERS
PADDLE MIXERS
IN-LINE MECHANICALLY DRIVEN MIXER
BATCH MIXER
REMOVEABLE PADDLE MIXER
DISPOSABLE PADDLE MIXER
BASICALLY LIMITED TO SMALL SHIPPING CONTAINERS (eg. - 55 GAL DRUMS)
BASICALLY LIMITED TO RELATIVELY SMALL SHIPPING CONTAINERS WITH SPECIFIC CONFIGURATIONS & FEATURES.
SMALL OR LARGE SHIPPING CONTAINERS OF VARIOUS CONFIGURATIONS & WEIGHTS (eg. SHIELDED OR UNSHIELDED) CAN BE USED.

FIGURE 12

FLOW DIAGRAM FOR

CEMENT INCORPORATION PROCESSES

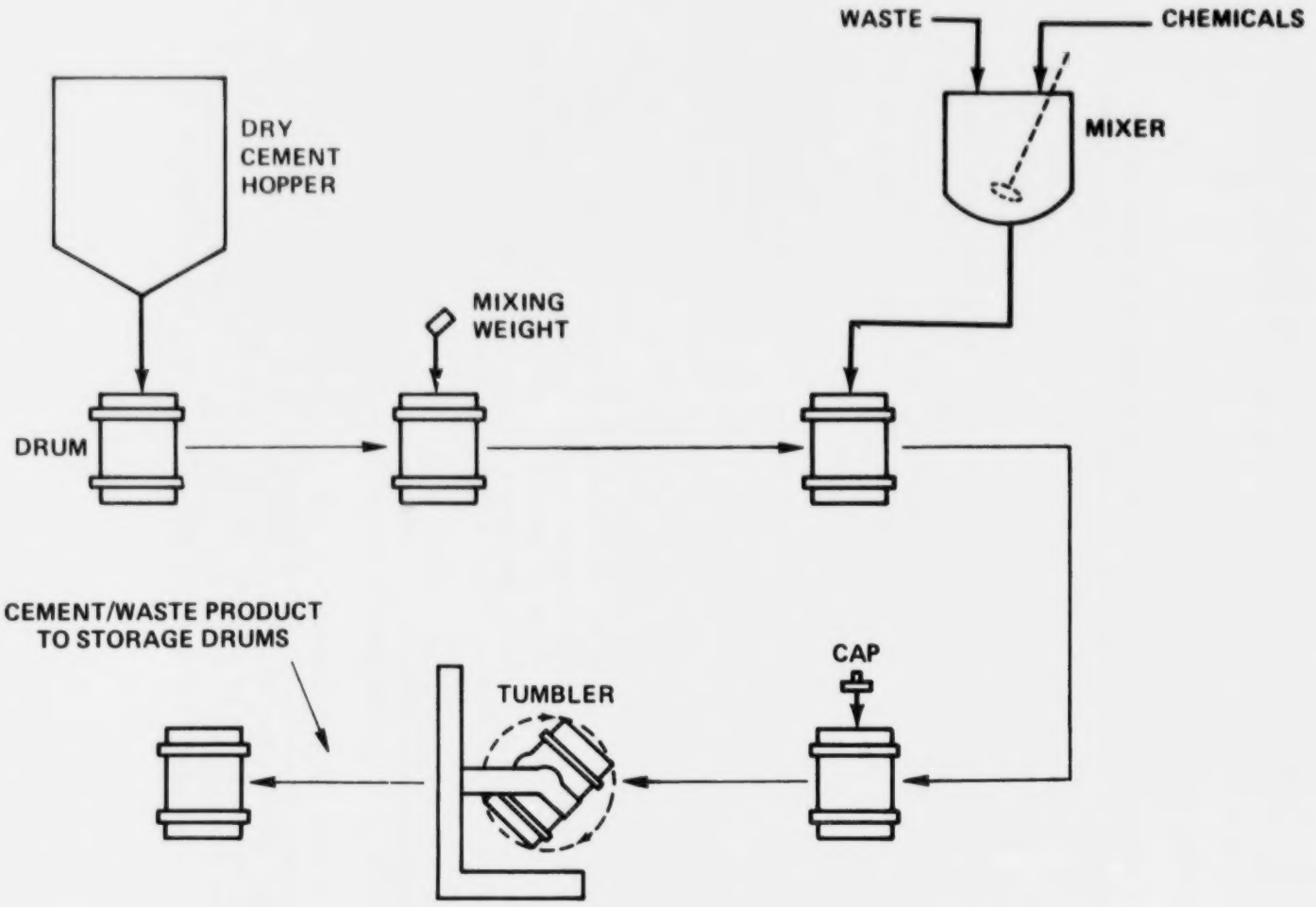

FIGURE 13

EXAMPLE OF IN-DRUM MIXING PROCESS

The present General Electric Solid Radwaste Systems use cement as the solidification agent with a disposable mixer and large disposal containers (2, 3, 38, 39).

2.2 External Mixer Process

This process technique involves both continuous in-line dynamic and static mixers and a batch mixing blade method (1-7). In-line dynamic mixing systems usually have small volume holdup times and utilize either ribbon mixers, pug mills, or positive displacement pumps. The waste and the cement are charged into one end of the mixer and the homogenous mix is discharged into a container and capped or sealed. The static mixer consists of a section of pipe containing stationary helical vanes which mixes the waste and cement as they flow through the mixer. The mixing blade system consists of introducing the liquid waste and cement into a conically shaped batch mixer. A mixing blade then blends the constituents and the product is drained into a container through the bottom of the mixer. Figure 14 illustrates this process. For example, ATCOR Inc. performs all its mixing with an in-line dynamic or mechanically driven mixer. The ATCOR process system mixes liquid and solid wastes with cement to produce a solidified product within a disposable receptacle. Basically, the system mixes separate feeds of moist radioactive waste or evaporator concentrates and dry cement in a small volume continuous mixer. Solid waste materials are preconditioned within the radioactive waste feed tank to provide sufficient moisture when mixed with the dry cement to achieve an acceptable cement mixture. The system not only solidifies resins, sludges and evaporator concentrates, but it can also be used to fix spent filter cartridges within a solidified matrix. In this case drums or large volume liners containing spent filter cartridges could be filled with a cement mixture that contains radioactive wastes. The cement waste mixture can be loaded directly into standard 208 liter drums or larger receptacles. Where waste is to be packaged in drums, drum capping and decontamination can also be provided. There is no preparation of drums required prior to filling.

The United Nuclear Industries (UNI) and Delaware Custom Material (DCM) systems for solidifying radwaste use an in-line batch mixer for waste and cement which is then mixed with sodium silicate in the shipping container. The UNI and DCM systems provide: (a) proportional pumps for metering waste feed; (b) in-line mixer to assure homogeneity; (c) single fill port for wastes; and (d) in-container solidification (3, 25, 26, 40, 41).

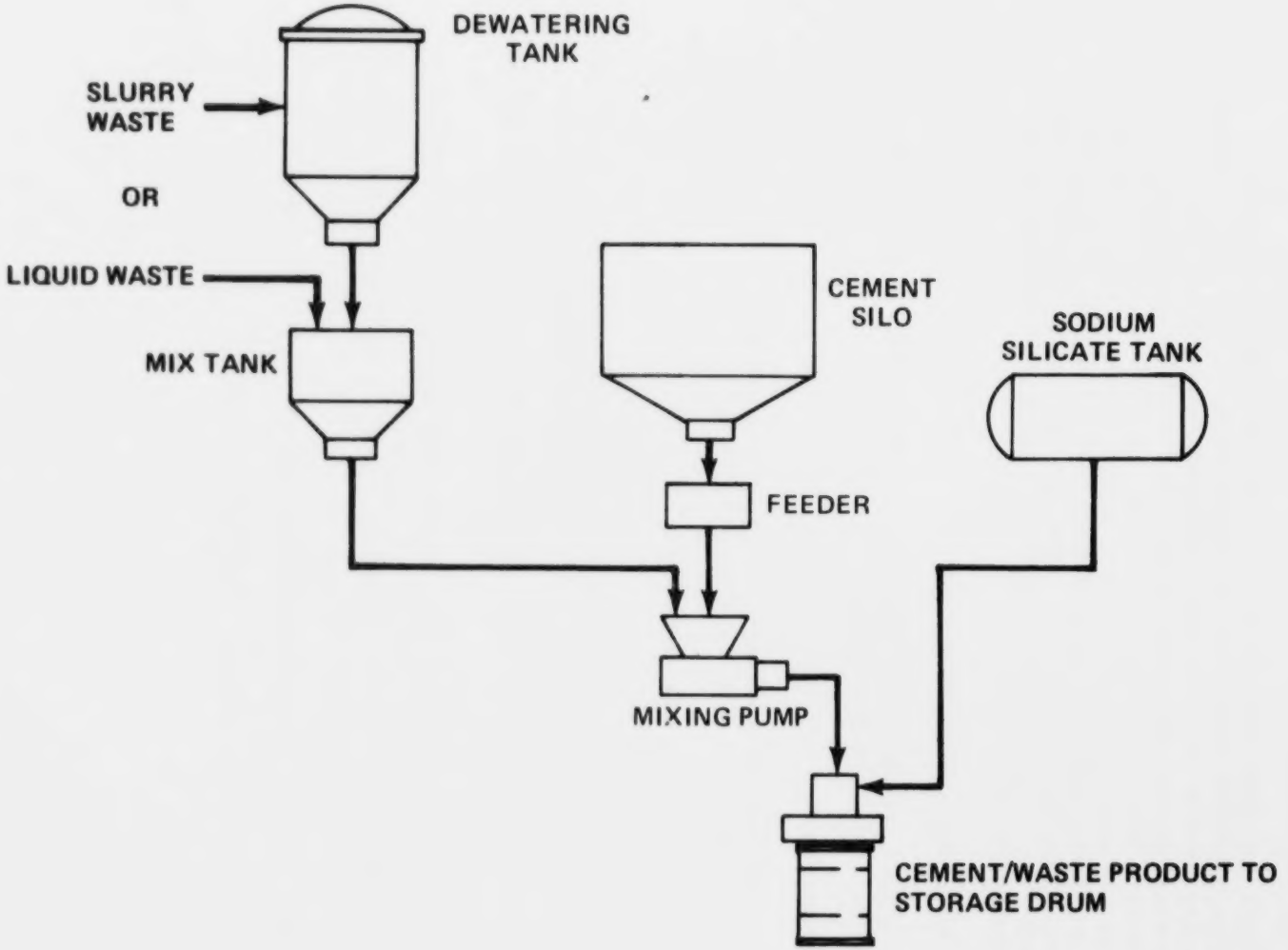

FIGURE 14

EXAMPLE OF EXTERNAL MIXING PROCESS

2.3 Other Processes

There are other variations of the two systems such as the Westinghouse Waste Encapsulation System which is basically a vacuum packaging process in which spent radioactive resins and waste evaporator bottoms are encapsulated using a cement vermiculite mixture in standard DOT 17 H drums (42). Also, the Delaware Custom Materials company utilizes the Chemfix, Inc., process which offers a complete service of equipment and chemicals for solidification of radioactive wastes, including a variety of inorganic and organic sludges. The process uses a combination of cements, shales and silicates as the solidification agent (26).

2.4 Advantanges and Disadvantages

The main reported advantages and disadvantages of using cement for insolubilization are as follows (1-9, 12):

Advantages

(a) No complex equipment; it is often possible to carry out the incorporation in the disposal receptacle.

(b) Low capital investment and low running costs; power requirements minimal.

(c) No applied heat required; low operating temperature means no fire risk and eliminates difficulties with off-gas purification.

(d) Most systems fully automatic; and therefore, operators can be trained easily.

(e) Waste-cement mixes are not grossly affected by pH.

(f) Cement is relatively cheap, but this is often off-set by the greater quantity required.

(g) Chemical and physical properties of cement are well known.

(h) Cement imparts good shielding properties.

(i) Natural alkalinity of cement is useful in helping to neutralize acidity in waste solutions.

(j) Little reported trouble with phase separation in the mix.

(k) Water is required for setting the mix so there is no need for extensive dewatering provided a satisfactory water/cement ratio is maintained.

(l) Presence of nitrates and nitrites and other oxidizing agents do not have the same detrimental effects as they can have when mixed with an organic material such as bitumen.

(m) Less subject to irradiation damage than bitumen.

Disadvantages

(a) The concentration of certain salts, such as borates, may cause the cement-waste matrix some difficulty in curing, and may cause deterioration and leaching over time at an abnormally high rate. However, progress is being made to solve this problem by using selected types of cement.

(b) The weight and volume of the final product will normally be as much as twice that for other corresponding solidification processes. The weight and volume increase is mainly due to the amount of cement which must be added to react with the residual water in the waste. (In ocean disposal, this is an advantage which overcomes the effect of buoyancy on the container volume.)

(c) If mixing equipment experiences operational trouble and break downs, this could require frequent cleaning of the equipment, particularly the blades.

(d) Nonautomated systems require several manual operations during the solidification process.

(e) Most studies have shown that when buried, and after the container rusts away, the cement will leach if in contact with ground water.

3.0 Polymeric Solidification Processes for Low-Level Radioactive Wastes

Incorporation of radioactive wastes into polymeric fixation agents is a relatively new solidification process when compared to incorporation in cement or bitumen. The solidification process can take place either at ambient temperatures or with hot evaporator concentrates (up to 60°C). Presently, several U.S. companies sell urea-formaldehyde (UF) solidification systems (as shown in Table I). All the organic processes are essentially batch processes where a catalyst is generally mixed with the wastes and polymer either in a premixer vessel or in the receptacle itself. The polymeric processes do not really solidify the wastes; the long chain molecules of the organic polymer are linked together to form a multi-voided sponge that "traps" the waste. (1-3, 12)

Paraffin and polyethylene based solidification agents can also be used to solidify wastes. These agents must be liquified by heating prior to mixing with the wastes (1, 2, 17, 43, 44).

The only industrial experience with polymeric solidification systems to date has been with the UF process. The process description, advantages and disadvantages are based on systems using UF, even though there are other organic polymer processes, such as the Dow Chemical and the Todd Research processes (45, 46), which are either not operational or have not been in operation long enough to provide operational information comparisons.

3.1 Urea Formaldehyde

The physical method of organic polymeric mixing depends upon the type of solidification agent and receptacle used. In general, there are three types of UF mixing:

1. In-container disposable paddle mixer.
2. In-line static mixer.
3. In-line mechanically driven mixer.

The in-container mixer is generally used for mixing resin beads with UF. The in-line dynamic mixer is used by UNI to mix liquid waste and UF prior to discharging into a receptacle. UF systems generally employ an in-line static mixer which contains stationary helical vanes to mix the fluids as they flow through the mixer. Just as the mixed polymer and waste are injected into a container, the acidic catalyst is added to initiate solidification. Figure 15 is a system diagram for UF indicating the incorporation steps. Figure 16 is a flow diagram of the static mixer technique.

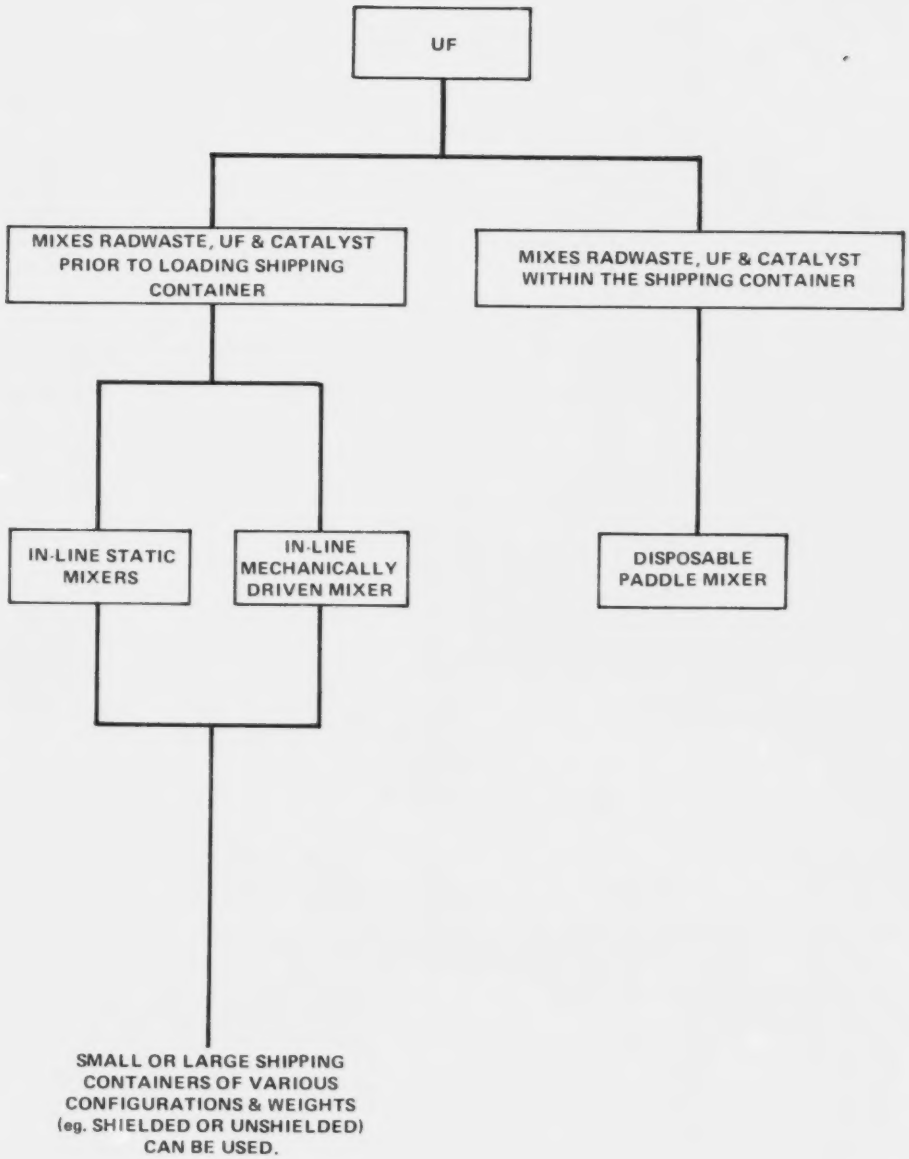
UF
MIXES RADWASTE, UF & CATALYST PRIOR TO LOADING SHIPPING CONTAINER
MIXES RADWASTE, UF & CATALYST WITHIN THE SHIPPING CONTAINER
IN-LINE STATIC MIXERS
IN-LINE MECHANICALLY DRIVEN MIXER
DISPOSABLE PADDLE MIXER
SMALL OR LARGE SHIPPING CONTAINERS OF VARIOUS CONFIGURATIONS & WEIGHTS (eg. SHIELDED OR UNSHIELDED) CAN BE USED.

FIGURE 15

FLOW DIAGRAM FOR UF INCORPORATION PROCESSES

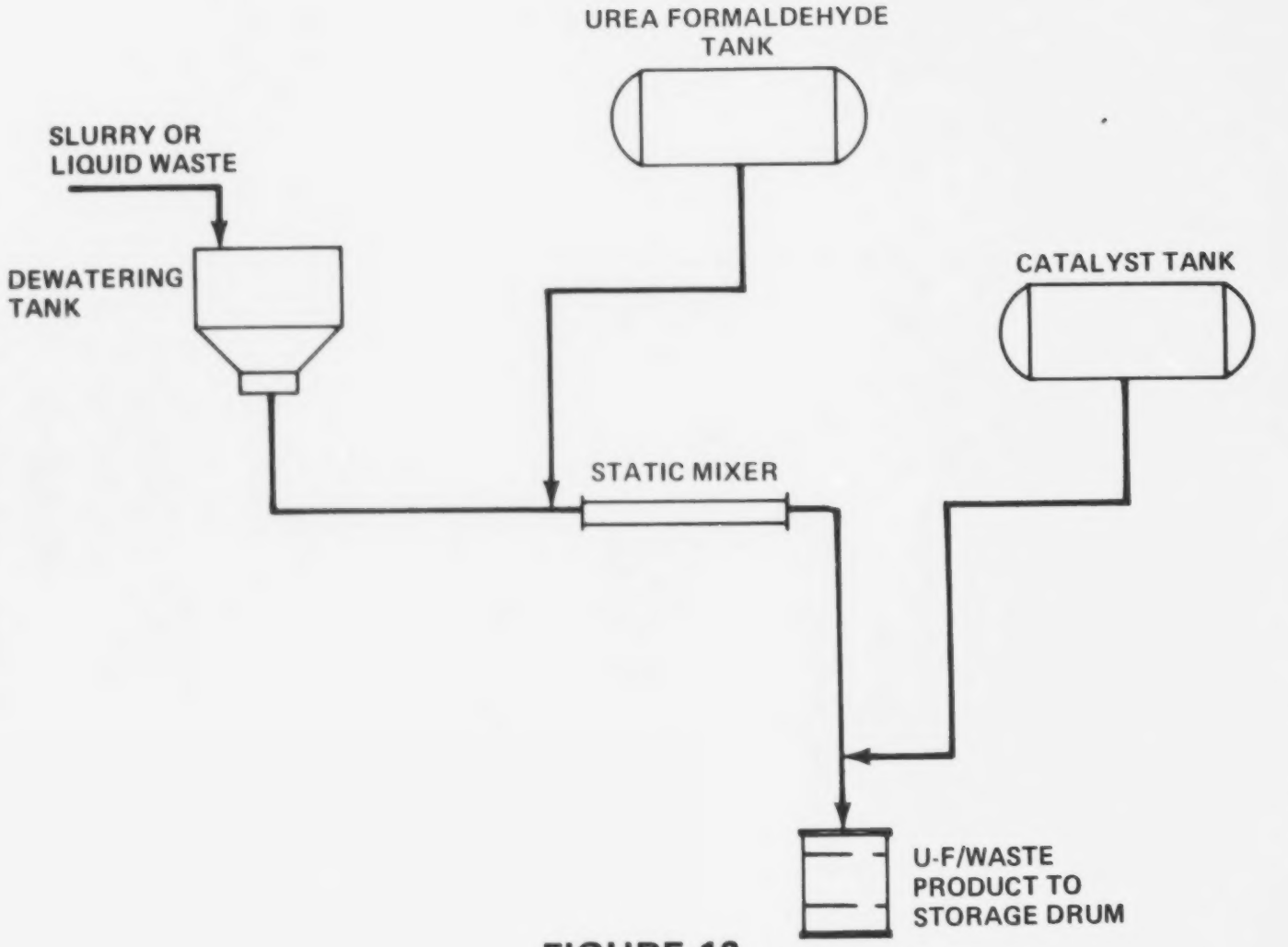

FIGURE 16

UREA FORMALDEHYDE INCORPORATION PROCESS — EXTERNAL MIXING

Protective Packaging, Inc. (PPI), a wholly owned subsidiary of Nuclear Engineering Co., developed and was the first company to design and sell, a system using a chemical solidification agent other then cement. Since then, they have filed several patent applications on the system and trademarked the name "TIGER-LOCK"[R]. Their patent applications cover both the use of the liquid solidification agent (TIGER-LOCK[R], a type of urea formaldehyde polymer), and also all the related hardware that makes up a TIGER-LOCK[R] Radwaste Solidification System. This includes the process equipment, control panel, power panel, and associated material handling equipment. The key aspects of the PPI design are: (a) three separate tanks, pumps, different size liners for radwaste, TIGER-LOCK[R], and catalyst respectively; (b) a premixer for the radwaste and UF to homogenize the slurry prior to contact with the catalyst; (c) a manual decoupling device to seal the liners that contain the cured waste; and (d) an automatic level detector to indicate filling to 90% volume (1-3, 47, 48).

TIGER LOCK[R] is a proprietary augmented agent of urea formaldehyde resin that is manufactured to strict physical and chemical specifications. The PPI system currently being sold includes TIGER LOCK[R] and catalyst (usually sodium bisulphate-$NaHSO_4$), associated processing equipment, and container liners for use in the transportation and burial of the solidified radwaste.

PPI suggests that the desired ratio of TIGER LOCK[R] to radwaste is 2:1 by volume. For this system the operator has the task of estimating the correct amount of catalyst for solidification which is highly dependent upon the quantity and type of radwaste that would be solidified by the PPI system.

Gel time of the product can be adjusted from minutes to hours by the catalyst concentration (normally about 1 to 3% by volume). If UF is used after its shelf life has been exceeded or at low temperatures or low viscosity, the "cottage cheese" effect will occur, i.e., little solidifying and essentially a settling of materials of different density within the container.

ANEFCO, Inc., offers a waste solidification system using urea formaldehyde and sulfuric acid, or an equivalent catalyst, as a solidifying agent. Their process system uses a 3785 liter batch tank, a static mixer and a disposable polyethylene liner in the disposal container (2, 3).

R-Registered Trademark, Protective Packaging, Inc., Jeffersontown, Kentucky

Hittman Nuclear & Development Corporation offers radioactive waste solidification systems using cement or a polymer such as urea formaldehyde as the solidifying agent. Chemical additives are used with both agents to enhance the efficiency, i.e., volume of waste per unit volume of solidified product. The disposable containers used to package radwaste vary in size from a standard 208 liter drum up to 5.7 m^3 capacity (2, 3, 21, 49).

The United Nuclear Industries also offers a radwaste solidification system utilizing urea formaldehyde (UF) as the solidification agent (1-3, 25, 40, 41). The use of the UF material permits the use of in-line static mixers with no moving parts. Solidification of the waste - UF mixture is accomplished using either a sodium bisulfate catalyst (pH range of 3 to 7.5) or a phosphoric acid catalyst (pH range of 3 to 10).

Chem-Nuclear Systems Inc., offers either portions of or a complete waste system design, component selection, procurement, fabrication, construction, installation and operation of solidification systems using either cement or urea formaldehyde as the solidification agent. Chem-Nuclear also has available a mobile solidification unit using the UF system (2, 3, 50).

Energy Incorporated and Newport News Industrial Corporation have jointly developed a radioactive waste reduction (designated RWR-1™) system, to convert low and medium level liquid and solid combustible radioactive wastes to solids by a fluidized bed calcining and incineration process. The RWR-1™ system reduces the volume of both liquid and solid radwastes, e.g., concentrated chemical wastes, filter sludges, spent resin beads, rags and other similar materials, and produces a granular, anhydrous solid which may be placed directly in burial containers or incorporated into matrices such as concrete, urea formaldehyde or bitumen for burial (2, 51). The RWR-1™ system has indicated volume reduction factors of 5-80:1, depending on the material; and the main components are designed for remote operation. The operating parameters include process temperatures of 400 C for the calciner and 1000 C for the incinerator, with waste feed rates of 45-90 Kg per hour and 132 liters per hour.

The Aerojet Energy Conversion Company has marketed a VR-20 Radioactive Waste Management System which reduces the volume and encapsulates the waste. The volume reduction is achieved by conversion of all liquid wastes into anhydrous calcined solids and drying of dewatered spent resins and sludges. These solid wastes can then be

encapsulated in cement, UF or bitumen as the solidification agent for subsequent shipment and burial. Using the VR-20 process, the volume of liquid waste could be reduced by a factor of 10-20, while the volume of liquid spent resins and sludges could be reduced by a factor of 2-4 when compared with other solidification methods.

The main feature of this system is a fluidized bed calciner which receives the radioactive liquid waste feed containing the dissolved chemical solids and processes these aqueous solutions into free-flowing anhydrous particles. Concentrated radioactive liquid waste (evaporator bottoms, etc.) is pumped from the concentrated liquid waste storage tank to a heated fluidized bed calciner concentrator. The volatiles exit with the water vapor at the top of the fluidized bed concentrator, leaving behind the dissolved solids. The granular solid produced can then be encapsulated (2, 3, 52-56).

3.2 Advantages and Disadvantages

The main reported advantages and disadvantages of using urea-formaldehyde for insolubilization are as follows (1-3, 9, 12).

Advantages

(a) The amount of waste capable of incorporation in a receptacle wih UF is about 30% by volume more than with cement.

(b) For shipping not requiring radiation shields, shipping cost with UF or polymerics is less than for cement and bitumen due to the ability to put more waste in a given container and a lower density for the mixture.

(c) Mixture of UF and radwaste are not combustible. Further, no detectable exothermic reaction occurs with UF.

Disadvantages

(a) For shipments requiring radiation shields, UF or polymeric solidified materials, due to its lower bulk density and higher activity, requires more shielding than materials solidified with cement.

(b) It appears that routine attainment of the complete elimination of free standing water is a problem with encapsulated UF radwaste, particularly those having lower concentrations of the polymer (volume ratios of 1 to 3 or less).

(c) According to the utility operators, it is difficult to work with UF because of the relatively low viscosity of the mixture, which permits settling or floating (segregation) of materials of different densities.

(d) Solidification time is affected by both the pH of the mixture, which is regulated by the amount of catalyst, and the temperature of the mixture.

(e) The UF shelf life is limited and is dependent upon storage conditions.

(f) Equipment must be designed to eliminate fume problems with UF; the odor is disagreeable even in small concentrations.

(g) Some manufacturers of UF have stated that this product is biodegradable; also the catalyst is corrosive to most metals.

(h) During the solidification process when the UF-radwaste mixture is exposed to air, water evaporates from the mass, but if the matrix remains in an air-tight container, the mixture will remain semi-liquid.

3.3 Other Polymeric Processes

Polyethylene agents are not used commercially in the U.S. Polyethylene is a superior solidifying agent for most organic liquid wastes. The waste is combined with molten polyethylene inside a heated chamber in which the water and other volatile constituents are evaporated. The mixed and dehydrated liquid product is discharged to a container where it solidifies upon cooling. The final product is a solid plastic block, which is relatively inert at room temperature and is insoluble in water. It has good freeze-thaw characteristics and a storage life of several years. Polyethylene is completely combustible and can be incinerated. It is flammable with a flash point of 250 C (1, 2, 17, 43, 44).

United Technologies-Chemical Systems Division offers the Inert Carrier Process which handles ingredients in an inert organic liquid. The operating concept is based on dispersal of the reactants in an inert carrier to provide maximum surface area for solid-liquid reaction mechanism. In addition, the process provides for a clean separation of the reaction product from the inert carrier. The waste materials are

low viscosity dispersions in an inert carrier. The inert carrier is a fluid selected so that neither the starting materials nor the products are soluble in it or chemically reactive with it. The process has particular advantages in operations which require (a) preparation of compositions which are too viscous to mix by ordinary methods; (b) extremely intimate mixing of solids with small quantities of liquids; (c) safe control of highly exothermic chemical reactions; or (d) a closed system and/or remote controlled processing of hazardous, toxic, or explosive materials (57).

Dow Chemical Company has developed a radwaste solidification system using organic polymers that produces a radwaste product free of liquid, reasonably hard, and free standing. The solidification system is usable for all anticipated chemical decontamination solvents and regular wastes from nuclear power stations. To simulate disposal conditions, Dow evaluated the solidification product for the following: (a) compressive strength; (b) temperature cycling; (c) radiation stability; (d) leachability; (e) impact testing; (f) heat exposure; and (g) free liquid.

To date, Dow has solidified the following simulated wastes in the laboratory and in 208 liter drums, containing, no detectable free liquid: (a) spent decontamination solutions at pH's of 3 to 5 and 9 to 10 with 40% solids; (b) filter aid and slurries, 90/10 by volume; (c) ion exchange resins, 90/10 by volume; (d) PWR evaporator bottoms with a pH of 2.5 and 7% solids; and (e) BWR evaporator bottoms with a pH of 10.6 and 6% solids. After casting, the drums solidified within one hour. The radwaste to solidification agent ratio is a least 1.25 to 1 and as high as 2.5 to 1. A field demonstration was carried out by successfully solidifying 3400 liters of radioactive decontamination solvent at a nuclear power plant (45).

Todd Research and Technical Division is marketing a solidification agent called SAFE-T-SET^{R}, which is a long chain linkage organic polymer. The agent can be used with concentrated low-level liquid radioactive wastes from filtration, precipitation, ion-exchange or evaporation. The set-up time varies from one minute to several hours depending on the amount used in proportion to the volume of waste. One-half kilogram of SAFE-T-SET^{R} will solidify 3.8 liters of liquid material. The agent can be tailored to any particular system or circumstances including pumping the waste and SAFE-T-SET^{R} mixture and can be adapted to molds of any type. The solidified matrix remains stable under conditions of freezing, high temperature and leaching (46).

R-Registered Trademark, Todd Research and Technical Division, Galveston, Texas.

4.0 Use of Absorbents for Solidification of Low-Level Radioactive Wastes

Absorbents are used to eliminate free standing liquids by virtue of their ability to hold water molecules within their pores. The absorbent is, however, not chemically bound to the waste, nor does it represent a free standing monolithic solid; therefore, the absorbents should not be considered as solidification agents. Further, they do not provide or enhance resistance to leaching, if water comes in contact with the absorbed radioactive materials. The absorbents are stored in a dry environment and are placed in the shipping container prior to adding radioactive liquids. Some commonly used absorbents are vermiculite, clays, silica gel, plaster of paris, microcell and/or diatomaceous earth filter aid.

Vermiculite, dehydrated clay granules, and diatomite absorbents have been routinely used for liquid wastes, with perhaps vermiculite the most widely used. The absorbent method, when properly applied will physically entrap the waste liquid so that no appreciable free liquid will leak out if the container is breached. However, in most cases, the liquids can be displaced readily by the addition of water.

In preparation, the receptable is filled with vermiculite and liquid waste equivalent to about 1/3 to 1/2 of the volume. For some materials such as the diatomaceous earth, physical mixing of the liquid and absorbent may be necessary. Care must be taken with all absorbent to avoid supersaturation (1, 2, 12).

5.0 Cited References

1. Alternatives for Managing Wastes from Reactors and Post Fission Operations in the LWR Fuel Cycle, Volume 2: Alternatives for Waste Treatment, Report No. ERDA-76-43, U. S. Energy Research and Development Administration, Washington, D. C. May 1976.

2. W. F. Holcomb and S. M. Goldberg, Available Methods of Solidification for Low-Level Radioactive Wastes in the United States, Report No. ORP/TAD-76-4, U. S. Environmental Protection Agency, Office of Radiation Programs, Washington, D. C., December 1976.

3. Radioactive Waste Management for Nuclear Power Reactors, UCLA Extension Course, Engineering 821.7, Los Angeles, California, October 20-23, 1975.

4. R. H. Burns, "Solidification of Low and Intermediate Level Wastes", Atomic Energy Review, 9 (3), 1971.

5. Treatment of Low and Intermediate-Level Radioactive Waste Concentrates, Technical Report Series No. 82, STI/DOC/10 82, International Atomic Energy Agency, Vienna, Austria 1968.

6. Management of Low and Intermediate-Level Radioactive, STI/PUB/264, International Atomic Energy Agency, Vienna, 1970.

7. Management of Radioactive Wastes at Nuclear Power Stations , Safety Series No. 28, International Atomic Energy Agency, Vienna 1968.

8. L. H. Barrett, "Solid Waste Treatment at Nuclear Stations", presented at 1975 Annual Conference, Southeastern Electric Exchange, April 17-18, 1975.

9. P. Columbo and R. M. Neilson, Jr., Critical Review of the Properties of Solidified Radioactive Waste Packages Generated at Nuclear Power Reactors, USNRC Report BNL-NUREG-50591, Brookhaven National Laboratory, Upton, New York, December 1976.

10. R. E. Blanco, H. W. Godbee, and E. J. Frederick, "Radioactive Wastes...Incorporating Industrial Wastes in Insoluble Media, "Chemical Engineering Progress, 66(2), pages 50-56, February 1970.

11. W. Hild, W. Kluger, and H. Krause, "Bituminization of Radioactive Wastes at the Nuclear Research Center," Transactions of ANS, Vol. 19, 1974 Winter meeting of the American Nuclear Society, Washington, D. C., October 27-31, 1974.

12. J. P. Duckworth, M. J. Jump, and B. E. Knight, Low-Level Radioactive Waste Management Research Project Final Report, Nuclear Fuel Services, Inc., West Valley, New York, September 15, 1974.

13. Bituminization of Radioactive Wastes, Technical Reports Series No. 116, International Atomic Energy Agency, Vienna, 1970.

14. J. E. Stewart, and R. Herter, "Solid Radwaste Experience in Europe Using Asphalt," ASME-IEEE Joint Power Generation Conference, Portland, Oregon, September 28-October 1, 1974.

15. Radioactive Residues, Their Origin and Elimination, KS-Information Brief Report No. 20, Werner and Pfleiderer Engineers, Stuttgart, Federal Republic of Germany, 1975.

16. G. Meier, and W. Bahr, The Incorporation of Radioactive Wastes into Bitumen, Part 1: The Bituminization Plant for Radioactive Evaporator Concentrates at the Karlsruhe National Research Center, Report KFK-2104, Karlsruhe, Federal Republic of Germany, April 1975.

17. G. L. Fitzgerald, H. W. Godbee, R. E. Blanco, and W. Davis, Jr., "The Feasibility of Incorporating Radioactive Wastes in Asphalt or Polyethylene," Nucl. Appl. Technol., Vol 9, p. 821, 1970.

18. Topical Report: Radwaste Volume Reduction and Solidification System, Report No. WPC-VRS-001, Werner & Pfleiderer Corporation, Waldwick, N. J., November 1976 (Revision 1-May 1978).

19. J. E. Stewart, "European Experience with Asphalt Packaging of Radioactive Wastes," presented at the Fifth International Symposium on Packaging and Transportation of Radioactive Materials, Las Vegas, Nevada, May 7-12, 1978.

20. R. J. Stouky, "Operating Costs Nuances of Nuclear Power Plant Radioactive Waste Disposal," presented at The Symposium on the Management of Low-Level Radioactive Waste, Atlanta, Georgia, May 23-27, 1977.

21. R. B. Wilson, "Low-Level Radwaste Packaging - Why Not Cement," presented at Fifth International Symposium Packaging and Transportation of Radioactive Materials, Las Vegas, Nevada, May 7-12, 1978.

22. W. F. Holcomb, Causes of Concrete Cracking and Methods of Control, USAEC Report NVO-38-26, Fenix and Scisson, Inc., Las Vegas, Nevada, July 1970.

23. Concrete Manual, 7th Edition, U. S. Department of Interior, Bureau of Reclamation, U. S. Government Printing Office, Washington, D. C., 1966.

24. S. Brunauer and L. E. Copeland, "The Chemistry of Concrete," Scientific American, p 80, April 1964.

25. W. H. Heacock, "Alternative Nuclear Waste Solidification Processes," Waste Management 75 Symposium, R. G. Post (Editor), University of Arizona, Tucson, Arizona, March 1975.

26. John Hays, Personal Communication, Delaware Custom Material, State College, Pennsylvania 1976.

27. Final Environmental Impact Statement, Management of Intermediate Level Radioactive Waste, Oak Ridge National Laboratory, Oak Ridge, Tennessee, Report ERDA-1553, U. S. Energy Research and Development Administration, Washington, D. C., September 1977.

28. Environmental Statement, Radioactive Waste Facilities, Oak Ridge National Laboratory, Oak Ridge, Tennessee, Report WASH-1532, U. S. Atomic Energy Commission, Washington, D. C., August 1974.

29. W. de Laguna, "Radioactive Waste Disposal By Hydraulic Fracturing", Industrial Water Engineering, page 32, October 1970.

30. J. G. Moore and E. W. McDaniel, "Fixation of Intermediate - Level Radioactive Waste in Hydrofracture Grout", presented at the 80th Annual Meeting of the American Ceramic Society, Detroit, Michigan, May 6-11, 1978.

31. P. Colombo and R. M. Neilson, Jr., "Some Techniques for the Solidification of Radioactive Wastes in Concrete", Nuclear Technology, Vol. 32, p. 30, January 1977.

32. Development of Durable Long-Term Radioactive Waste Composite Materials, USAEC Progress Reports No. 1-10, Brookhaven National Laboratory, Upton, N.Y., July 1972-April 1975.

33. L. E. Kukacka, "Production Methods and Applications For Concrete Polymer Materials," Presented at the 70th Annual Meeting of the American Institute of Chemical Engineers, New York, November 13-17, 1977.

34. M. Steinberg, et al, "Concrete-Polymer Materials Development," Nuclear News, p. 48, September 1970.

35. Draft Environmental Impact Statement, Las Alamos, Scientific Laboratory Site, Las Alamos, New Mexico, Report DOE-EIS-0018 D, U. S. Department of Energy, Washington, D. C., May 1978.

36. R. E. McLin, et al, "Experience with Cement Fixation of Nuclear Waste at LASL," presented at the 80th Annual Meeting of the American Ceramic Society, Detroit, Michigan, May 6-11, 1978.

37. J. Stock, "A Radwaste Disposal System Type VI", Stock Equipment Company, Cleveland, Ohio, May 1972.

38. J. M. Smith, and J. E. Kjemtrup, "BWR Development in Nuclear Plant Effluent Management," presented at American Power Conference, Illinois Institute of Technology, Chicago, Illinois, April 18-20, 1972.

39. H. L. Loy and D. C. Saxena, "Processing and Packaging of Solid Wastes From BWR's," Proceedings 3rd International Symposium, Packaging and Transportation of Radioactive Materials, Richland, Washington, August 16-20, 1971, Report CONF 710801 (Vol 1), U. S. Atomic Energy Commission.

40. Summary of Qualifications for Furnishing of Nuclear Power Plant Radwaste Solidifixation Systems, Douglas United Nuclear, Inc., Richland Washington, January 1972

41. Radwaste Solidification Septems, United Nuclear Industries, Inc., 1975

42. Personal Communication, Westinghouse Electric Corporation (PWR Systems Division) Pittsburg, Pennsylvania, 1976.

43. R. V. Subramanian, and N. Raff, "Polymeric Immobilization of Low-Level Radioactive Wastes," presented at American Institute of Chemical Engineers 80th National Meeting, Boston, Mass., September 7-10, 1975.

44. R. V. Subramanian and R. Mahalingam, Immobilization of Hazardous Residuals by Encapsulation, Annual Letter Technical Report to National Science Foundation for Grant AEN-75-06583, Washington State University, Pullman, Washington, April 30, 1976.

45. H. E. Filter, The Dow System for Solidification of Low-Level Radioactive Waste from Nuclear Power Plants, The Dow Chemical Company, Midland, Michigan, October 1976.

46. The Use of "SAFE-T-SET" As a Radioactive Liquid Waste Solidification Medium, Todd Shipyards Corporation, Galveston, Texas, May 1967.

47. K. A. Gablin, and J. H. Leonard, "Leachability Evaluation of Radwastes Solidified with Various Agents," The American Society of Mechanical Engineers, United Engineering Center, New York, N.Y., 74-WA/NE-8, 1974.

48. S. K. Cosgrove, K. M. Emmerich, and J. H. Leonard, Interim Report on Evaluation of Solidification Techniques for Low-Level Nuclear Waste Materials, J. H. Leonard Associates, Cincinnati, Ohio, August 1974.

49. P. T. Tuite, S. R. Zimmerman, and G. K. Bolat, "A System for Solidification and Packaging of Radioactive Waste at a PWR," Presented at the American Power Conference, Illinois Institute of Technology, Chicago, Illinois 1972.

50. C. D. Johnson, "Modern Radwaste System: An Overview," Chemical Engineering Progress, Vol. 72, No. 3, page 43, March 1976.

51. (a) Energy Incorporated Sales Brochure, "RWR-1: Radioactive Waste Reduction," Energy Inc., Idaho Falls, Idaho, 1975;
(b) Newport News Industrial Corporation Sales Brochure,"Radwaste Volume Reduction, RWR-1™," Newport News Industrial Corp., Newport News, Virginia, 1976;
(c) News Release Technical Bulletin, "RWR-1™," Newport News Industrial Corp., Newport News, Virginia, October 1, 1976.

52. L. E. White, and R. Garcia, "Use and Economic Advantages of Fluid Bed Calciners for Volume Reduction of Liquid Radwaste," The American Society of Mechanical Engineers for presentation at the Winter Annual Meeting, New York, New York, November 17-22, 1974.

53. L. E. White and R. Garcia, "Environmental Survey of Transportation of Radioactive Wastes to the Burial Site," Fourth National Symposium on Radioecology, Oregon State University, May 12-14, 1975.

54. L. E. White and R. Garcia, "Environmental Impact of Radioactive Waste Solidification Process on Burial Site Operations," Fourth National Symposium on Radioecology, Oregon State University, May 12-14, 1975.

55. L. E. Tokerud and R. Garcia, "Iodine Decontamination Factor for Liquid Radioactive Waste Volume Reduction System," Trans. Am. Nucl. Soc., 23, 264, 1976.

56 Aerojet Energy Conversion Company, Topical Report: Fluid Bed Dryer, Report No. AECC-1-A, Sacramento, California, February 21, 1975.

57. Randall D. Sheeline, Personal Communication, United Technologies, Sunnyvale, California, September 1976.

6.0 Additional References

1. S. E. Pihlajavaara and J-P. Aittola, "Radwaste Concrete: Solidification of Nuclear Wastes with Portland Cement," presented at the 80th Annual Meeting of the American Ceramic Society, May 6-11, 1978, Detroit, Michigan.

2. H. E. Flora, "Radioactive Waste Processing for Nuclear Power Plants," presented at the 11th Annual Liberty Bell Corrosion Course, September 13, 1973, Philadelphia, Pennsylvania.

3. Draft Report: The Conditioning of Residues from the Treatment of Low- and Intermediate-Level Radioactive Wastes and Criteria for Their Storage or Disposal on Land, International Atomic Energy Agency, Moscow, USSR, December 9-13, 1974.

4. T. B. Mullarkey et al (NUS Corporation), A Survey and Evaluation of Handling and Disposing of Solid Low-Level Nuclear Fuel Cycle Wastes, Report AIF/NESP-008, Atomic Industiral Forum, Inc., Washington, D. C., October 1976.

5. P. Colombo and R. M. Neilson, Jr., Properties of Radioactive Wastes and Waste Containers Progress Reports:

BNL-NUREG-50571	April-June 1976
BNL-NUREG-50617	July-September 1976
BNL-NUREG-50664	October-December 1976
BNL-NUREG-50692	January-March 1977
BNL-NUREG-50763	April-June 1977
BNL-NUREG-50774	July-September 1977
BNL-NUREG-50837	October-December 1977

Brookhaven National Laboratory, Upton, New York.

6. H. L. Freese and W. T. Gregory III, "Volume Reduction of Liquid Radioactive Wastes Using Mechanically Agitated Thin-Film Evaporators," presented at the 85th National Meeting of the American Institute of Chemical Engineers, June 4-8, 1978, Philadelphia, Pennsylvania.

7. A. H. Kibbey and H. W. Godbee, "Solid-Radioactive-Waste Practices at Nuclear Power Plants," Nuclear Safety, Vol 16, No. 5, p. 581, September-October 1975.

V. LEACHING STUDIES

The most likely mechanism of radionuclide release to the surroundings is by solution in the water existing in the environs of the burial site. Therefore, measurements are usually attempted to indicate the rate at which radionuclides are leached from the solidified products. The leachability properties of a radwaste solidification matrix will strongly influence the amount of treatment, containment and surveillance that will be required.

A review of the various leach rate tests and results indicates a wide variety of testing methods in use by industry and government facilities with little attempt at standardized techniques. In addition, the tests are not performed to simulate actual ground media and leachate conditions available at the various facilities and potential sites around the U. S. This along with the many other factors involved, e.g., radionuclide measured, pH, temperature, leachant composition, have resulted in different test procedures and results reported in many different units. Also, in connection with leaching, consideration must be given to the corrosion of the various containers, both internally and externally. Then there is also the deterioration of the matrices by such things as radiation and bacteriological attack.

As a result many studies have been made on leach rates and several studies are still underway. The International Atomic Energy Agency has proposed a leach test method (1) which is presently under evaluation, in a modified form, as a standardized leach test procedure by the Brookhaven National Laboratory (2,3) for low-level solidified radioactive wastes.

Studies have been underway for many years to provide information on the leach resistance of various solidified waste products (4-16). Leach rates for alkali and alkaline earth, rare earth, and actinide elements from various waste matrices are compared in Table III (17). A comparison shows: (a) cement has wide ranging leach rates; (b) calcines are extremely leachable; (c) that for a given waste matrix the leach rates for rare-earth and actinide elements are about a factor of 1,000 less than those for alkali and alkaline earth elements; and (d) the leach rates for rare-earth and actinide elements from cements and grouts are as low as those from glasses. Additional leachability work needs to be accomplished in the near future to permit a more complete understanding of the environmental impact of both high- and low-level solidified radioactive waste.

Table III

COMPARISONS OF LEACH RATES FOR VARIOUS SOLIDIFIED WASTE PRODUCTS (17)

Leach Rates, Grams/Cm^2-Day

	Alkali and Alkaline-Earth	Rare Earth	Actinide
Calcines	10^{-1}-1	10^{-4}-10^{-3}	--
Ceramics			
Phosphate	10^{-5}-10^{-2}	10^{-9}-10^{-6}	--
Devitrified Phosphate glass	10^{-4}-10^{-2}	--	--
Glasses			
Borosilicate	10^{-7}-10^{-5}	10^{-9}-10^{-7}	--
Phosphate	10^{-8}-10^{-5}	10^{-9}-10^{-6}	--
Aluminosilicate	10^{-8}-10^{-7}	--	--
Bitumens	10^{-7}-10^{-4}	--	10^{-8}-10^{-7}
Cements	10^{-6}-10^{-1}	--	10^{-9}-10^{-7}
Grouts	10^{-7}-10^{-4}	--	10^{-7}

1.0 References

1. E. D. Hespe (Ed), "Leach Testing of Immobilized Radioactive Waste Solids," Atomic Energy Review, Vol. 9, No. 1, 1971.

2. P. Colombo and R. M. Neilson, Jr., Critical Review of the Properties of Solidified Radioactive Waste Packages Generated at Nuclear Power Reactors, Report BNL-NUREG-50591, Brookhaven National Laboratory, Upton, New York, December 1976.

3. P. Colombo and R. M. Neilson, Jr., Properties of Radioactive Wastes and Waste Containers, Quarterly Progress Report, April-June 1976, Report BNL-NUREG-50571, Brookhaven National Laboratory, Upton, New York, October 1976.

4. J. E. Mendel, A Review of Leaching Test Methods and the Leachability of Various Solid Media Containing Radioactive Waste, USAEC Report BNWL-1765, Battelle Pacific Northwest Laboratories, Richland, Washington, July 1973.

5. J. G. Moore, et al, Development of Cementitious Grouts for the Incorporation of Radioactive Wastes, Part 1: Leach Studies, USERDA Report ORNL-4962 April 1975 and Part 2: Continuation of Cesium and Strontium Leach Studies, USERDA Report ORNL-5142 September 1976, Oak Ridge National Laboratory, Oak Ridge, Tennessee.

6. J. G. Moore, et al, "Leach Behavior of Hydrofracture Grout Incorporating Radioactive Wastes," Nuclear Technology, Vol 32, p. 39, January 1977.

7. H. W. Godbee and D. S. Joy, Assessment of the Loss of Radioactive Isotopes From Waste Solids to the Environment, Part 1: Background and Theory, USAEC Report ORNL-TM-4333, Oak Ridge National Laboratory, Oak Ridge, Tennessee, February 1974.

8. J. A. Kelley and R. M. Wallace, "Procedure for Determining Leachabilities of Radioactive Waste Forms," Nuclear Technology, Vol. 30, p. 47, July 1976.

9. W. F. Merritt, "High-Level Waste Glass: Field Leach Test," Nuclear Technology, Vol. 32, p. 88, January 1977.

10. J. A. Kelley and W. N. Rankin, Correlation of Radionuclide Leachabilities with Microstructures of Glasses Containing Savannah River Plant Waste, USERDA Report DP-1411, Savannah River Laboratory, Aiken, South Carolina, May 1976.

11. M. W. Wilding and D. W. Rhodes, Leachability of Zirconia Calcine Produced in the Idaho Waste Calcining Facility, USAEC Report IN-1298, National Reactor Testing Station, Idaho Falls, Idaho, June 1969.

12. B. E. Paige, Leachability of Alumina Calcine Produced in the Idaho Waste Calcining Facility, USAEC Report IN-1011, National Reactor Testing Station, Idaho Falls, Idaho, July 1966.

13. R. H. Burns, "Solidification of Low- and Intermediate-Level Wastes," Atomic Energy Review, Vol. 9, No. 3, 1971.

14. J. H. Leonard and K. A. Gablin "Leachability Evaluation of Radwaste Solidified with Various Agents," 74-WA/NE-8, The American Society of Mechanical Engineers, New York, 1974.

15. S. L. Cosgrove, et al, Interim Report on Evaluation of Solidification Techniques For Low-Level Nuclear Waste Materials, J. H. Leonard Associates, Cincinnati, Ohio, August 1974.

16. H. E. Filter, The Dow System For Solidification of Low-Level Radioactive Wastes From Nuclear Power Plants, The Dow Chemical Company, Midland, Michigan, October 1976.

17. Alternatives For Managing Wastes From Reactors And Post-Fission Operations In The LWR Fuel Cycle., Volume 2: Alternatives For Waste Treatment, Report ERDA-76-43 (Vol. 2 of 5), U. S. Energy Research and Development Administration, Washington, D. C., May 1976.

VI. CONCLUSIONS

In the foregoing sections brief descriptions have been presented of several established and proposed processes for the solidification of high- and low-level radioactive wastes and the glassification of high-level wastes. Each of the processes, as well as each of the solidified waste products, have a number of advantages and disadvantages.

The safe and final disposition of radioactive wastes produced in the nuclear fuel cycle is dependent on proper management, which includes the solidification of these wastes. Solidification is desirable to provide protection against dispersal in adverse events. Thermal and chemical stability, insolubility, and capability to withstand impact are the major advantages.

The importance of the solidification processes is seen in the emphasis put on the development not only in the U.S., but in other countries as well. In the development of criteria, standards, and regulations, the Environmental Protection Agency, the Nuclear Regulatory Commission, and the Department of Energy have recognized the need for containment and for solidification as a necessary requirement (1-4). Also, the National Academy of Sciences/National Research Council Panel on Waste Solidification recognized, in its 1978 Report on Solidification of High-Level Radioactive Wastes, that the first requirement of the solid waste form, is that it be capable of furnishing a major barrier to the migration of radionuclides from the waste.

As these processes and techniques receive more attention and regulatory requirements surface, the best combinations and options will also surface. This implies that further improvements are possible and that the limitations and potential problems have not necessarily been identified.

1.0 REFERENCES

1. J.J. Cohen, et al; Determination of Performance Criteria for High-Level Solidified Nuclear Waste, USNRC Report NUREG-0279, Lawrence Livermore Laboratory, Livermore, California, July 1977.

2. Background Report: Considerations of Environmental Protection Criteria for Radioactive Waste, U.S. Environmental Protection Agency, Office of Radiation Programs, Washington, D.C., February 1978.

3. E. L. Moore, Interim Report: Commercial Waste Packaging Studies, Task 2, High-Level Waste Package Acceptance Criteria Study, USDOE Report RHO-ST-10, Rockwell Hanford Operations, Richland, Washington, March 1978.

4. P. Colombo and R. M. Neilson, Jr., Critical Review of the Properties of Solidified Radioactive Waste Packages Generated at Nuclear Power Reactors, Report BNL-NUREG-50591, Brookhaven National Laboratory, Upton, New York, December 1976.

APPENDIX A

Presentation by William F. Holcomb, Office of Radiation Programs, Environmental Protection Agency Before the Panel on Waste Solidification Formed by the Committee on Radioactive Waste Management, under the Sponsorship of the National Academy of Sciences/National Research Council - February 1, 1977

I have been asked to present a few points concerning EPA's role in Radioactive Waste Management and our interest in waste solidification.

First, from the environmental protection standpoint, EPA is concerned with proper management of all types of radioactive wastes. Second, our objective is one of containment rather than planned release and dispersion. Third, EPA recognizes the need for environmental criteria and guidance for waste management. Fourth, EPA has been directed to establish environmental radiation protection standards for the terminal storage of high-level radioactive waste by 1978, as a result of the review by the Presidential Nuclear Policy Task Force, which led to the Presidential Nuclear Policy Statement of October 27, 1976. We are implementing a program to carry out these objectives. Consideration will be given to the many key issues involved in making these decisions. Philsophically, EPA's goal for the management of radioactive wastes is to assure that no unwarranted risks are imposed upon present or future generations.

Pragmatically, EPA believes this goal can be achieved, through the combined use of techniques for waste processing, containerization, engineering controls in site selection, construction and operation; in judicious use of carefully selected geologic environmental barriers; and in pre-planned emergency response procedures. The intent of this multi-faceted approach is to contain radioactive wastes within the earth until they have decayed to innocuous levels; and to protect cost-effectively, the environment and public health from any uncontrollable routine or accidental impacts, now and in the future.

The Federal Government recognizes that efficient solutions to radioactive waste disposal problems will require close coordination and cooperation among all agencies involved. In this regard, EPA has worked with NRC, ERDA,* USGS, CEQ, FEA,* ERC,* and OMB to assure the development of coordinated radioactive waste disposal plans. To

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* Now part of the Department of Energy

fulfill EPA's obligations we have developed a three-step waste management program. The first step involves the development of pertinent environmental criteria. The second step leads to the establishment of generally applicable environmental standards. The third step consists of the development of standards and regulations in areas where EPA has specific regulatory authority; such as under the Ocean Dumping Act, the Federal Water Pollution Control Act, and the Safe Drinking Water Act.* In carrying out these steps, EPA's objective is to assure adequate radiation protection for public health and the environment over the period of time that radioactive wastes remain a potential hazard. In all three steps, the requirements of the National Environmental Policy Act of 1969 (NEPA) will be followed; and full participation by all interested parties will be afforded in public hearings.

Generally Applicable Environmental Standards for High-Level Waste

The work EPA has planned in the preparation of quantitative, generally applicable, high-level radioactive waste disposal standards includes broad participation by Government, public and private organizations, as specified in NEPA. Also, these standards will not be for regulation of specific radwaste management practices, facilities, or sites, (which is NRC's responsibility); instead, they will consist of numercial standards to define present and future containment levels that will be protective of the environment.

In order to provide technical support for these environmental standards, EPA has established a priority project as a major focal point to help quantify potential long-term environmental impacts from high-level waste. This project will center on the following five areas:

1. The quantification of the high-level waste problem (i.e., the magnitude of the source term), and comparison with other categories of radioactive wastes;

2. The projected performance of engineering barriers (such as solidified waste matrices and packaging containers);

3. Assessment of the effectiveness of environmental barriers to mitigate radionuclide transport through geological strata and formations;

*Additionally the Clean Air Act, as amended August 1977.

4. Assessment of the potential risk and resultant impacts resulting from accidental releases;

5. Compilation and evaluation of all factors involved in implementing high-level numerical standards.

In carrying out steps 1 through 3, EPA does not intend to undertake independent engineering programs, but to use available information from industry, ERDA, NRC, USGS, and other sources to the maximum extent possible.

Due to the critical need for the timely completion of these tasks, EPA has established a condensed time schedule to compile, analyze, and publish the technical support information from this contract.

As noted in the above project areas, we are particularly interested in the effectiveness of engineering barriers. The purpose of this task is to assess both available and future technologies which provide engineering controls to high-level waste. There are already several programs both in this country and abroad which are mature and are being vigorously pursued. However, the commercial application is still in its infancy. In addition, we must look at the capabilities of the spent fuel itself for retaining waste for the so called throw away option.

We intend to assess the current process development programs in the U. S., U. K., Germany and France concerning (a) solidification systems for aqueous raffinate wastes, including such processes as calcination, vitrification, sintering, pelletization, and encapsulation within a metal matrix; (b) encapsulation of transuranic contaminated wastes; and (c) container utilization for use at potential terminal disposal sites. Calcination and vitrification systems have been developed to an industrial stage in France and Germany, and; therefore, the capabilities of these foreign technologies will be considered.

It is our intent to rely on the information from ERDA and NRC regarding the prime candidates for the solidification matrix processes for high-level wastes. We want to emphasize the need for early and frequent information interchange. It is our belief that the matrix and container to be used is a important and fundamental part of the disposal process. One should keep in mind that a reliable matrix

and container will provide added assurance during the early years (i.e., the first 20-30 years) of the repositories validation phase, that in the event of any problems arising, the container and/or matrix will still be retrievable.

The application of engineering controls, in terms of packaging, i.e., solidification, has always been part of EPA's development plan for attaining the necessary information for preparing the generally applicable environmental standards.

APPENDIX B

PRESIDENT CARTER'S STATEMENT ON NUCLEAR POWER POLICY
April 7, 1977

There is no dilemma today more difficult to resolve than that connected with the use of nuclear power. Many countries see nuclear power as the only real opportunity, at least in this century, to reduce the dependence of their economic well-being on foreign oil - an energy source of uncertain availability, growing price, and ultimate exhaustion. The U. S., by contrast, has a major domestic energy source - coal - but its use is not without penalties and our plans also call for the use of nuclear power as a share in our energy production.

The benefits of nuclear power are thus very real and practical. But a serious risk accompanies world-wide use of nuclear power - the risk that components of the nuclear power process will be turned to providing atomic weapons.

We took an important step in reducing the risk of expanding possession of atomic weapons through the Non-Proliferation Treaty, whereby more than 100 nations have agreed not to develop such explosives. But we must go further. The U. S. is deeply concerned about the consequences for all nations of a further spread of nuclear weapons or explosive capabilities. We believe that these risks would be vastly increased by the further spread of sensitive technologies which entail direct access to plutonium, highly enriched uranium or other weapons usable material. The question I have had under review from my first day in office is how can that be accomplished without foregoing the tangible benefits of nuclear power.

We are now completing an extremely thorough review of all the issues that bear on the use of nuclear power. We have concluded that the serious consequences of proliferation and direct implications for peace and security - as well as strong scientific and economic evidence - require

- a major change in U. S. domestic nuclear energy policies and programs; and
- a concerted effort among all nations to find better answers to the problems and risks accompanying the increased use of nuclear power.

I am announcing today some of my decisions resulting from that review.

First, we will defer indefinitely the commercial reprocessing and recycling of the plutonium produced in the U. S. nuclear power programs. From our own experience we have concluded that a viable and economic nuclear power program can be sustained without such reprocessing and recycling. The plant at Barnwell, South Carolina, will receive neither federal encouragement nor funding for its completion as reprocessing facility.

Second, we will restructure the U. S. breeder reactor program to give greater priority to alternative designs of the breeder, and to defer the date when breeder reactors would be put into commercial use.

Third, we will redirect funding of U. S. nuclear research and development programs to accelerate our research into alternative nuclear fuel cycles which do not involve direct access to materials useable in nuclear weapons.

Fourth, we will increase U. S. production capacity for enriched uranium to provide adequate and timely supply of nuclear fuels for domestic and foreign needs.

Fifth, we will propose the necessary legislative steps to permit the U. S. to offer nuclear fuel supply contracts and guarantee delivery of such nuclear fuel to other countries.

Sixth, we will continue to embargo the export of equipment or technology that would permit uranium enrichment and chemical reprocessing.

Seventh, we will continue discussions with supplying and recipient countries alike, of a wide range of international approaches and frameworks that will permit all nations to achieve their energy objectives while reducing the spread of nuclear explosive capability. Among other things, we will explore the establishment of an international nuclear fuel cycle evaluation program aimed at developing alternative fuel cycles and a variety of international and U. S. measures to assure access to nuclear fuel supplies and spent fuel storage for nations sharing common non-proliferation objectives.

We will continue to consult very closely with a number of governments regarding the most desirable multilateral and bilateral arrangements for assuming that nuclear energy is creatively harnessed for peaceful economic purposes. Our intent to is develop wide international cooperation in regard to this vital issue through systematic and thorough interntional consultations.

APPENDIX C

STATUS OF FUEL REPROCESSING IN THE U. S.

There are more than 90 million gallons of high-level radioactive wastes from Federal defense reprocessing plants presently in storage in the United States. Although some of this material has been reduced to solids, the bulk remains in liquid form. Most of this material, mainly the byproduct of nuclear weapons production and related research, is stored by the Department of Energy (DOE) at its Hanford Reservation in Washington, the Savannah River Plant in South Carolina, and the Idaho National Engineering Laboratory (INEL) in Idaho (1, 2).

Also in storage are about 600,000 gallons of high-level wastes from the reprocessing of fuels used in the operation of commercial nuclear plants. This waste is stored at the Nuclear Fuel Services' (NFS) spent fuel reprocessing plant near West Valley, New York. The NFS plant, after operating for six years, was shut down in 1972 to expand from a capacity of 300 to 750 metric tons per year and to modernize, bringing the plant to current standards in all respects. However, because of the substantial scope of the changes in regulatory requirements NFS in September 1976 announced that they no longer intended to reprocess fuel at the West Valley Plant (3, 4).

The General Electric Company built and tested the 300 metric tons per year capacity Midwest Fuel Recovery Plant at Morris, Illinois; but in 1974 the plant experienced technical problesm in starting up and was not operable in its current configuration. The plant did not reprocess any spent fuel and has never been licensed to operate as a reprocessing plant before being shutdown. A final decision regarding the plant remains to be made (5-8).

Allied-General Nuclear Services (AGNS) has under construction a plant at Barnwell, South Carolina. The AGNS plant is designed to reprocess 1600 metric tons per year using the conventional Purex process. The construction permit for this plant was granted in 1970 with the central part of the plant, the separations facility completed.

However, completion and licensing of the plant is now delayed by a complex situation including the general consideration of plutonium recycle and the deferral of commercial reprocessing in the U. S. The plant still lacks the facilities needed for the solidification of high-level waste and plutonium nitrate. Even the existing separations facility cannot be licensed to operate until the plutonium recycle

issue is resolved. Meanwhile, licensing reviews and hearings on safety and environmental issues are continuing (5-8).

Finally, Exxon Nuclear Company, Inc., has filed a partial application in connection with their plans to construct and operate a fuel reprocessing plant in the Oak Ridge, Tennessee area. The Exxon plant design has an initial capacity of 1500 metric tons per year with a growth capacity to 2100 metric tons per year. The plant would use the conventional Purex process for reprocessing low enriched uranium oxide fuel. Safety and environmental reviews have been by NRC, but a construction permit cannot be granted until the plutonium recycle question is resolved (5-9).

The volumes of radioactive wastes will continue to grow as nuclear power production increases in the years ahead. If commercial spent fuel reprocessing were allowed, high-level radioactive wastes generated from these operations would require storage, reduction in volume, and finally disposal. These are activities that are under continuing investigation. They have not been fully resolved. Because of the relatively long life of many of the radioactive materials involved, isolation of these wastes from the environment will be required for many thousands of years.

The current approach to the management of high-level radioactive waste has two principal steps: (1) converting these wastes to a less mobile, solid form, thereby reducing the potential for accidental leakage to the environment, and (2) finding a suitable place to put them that will provide adequate isolation and protection to the public for extremely long periods of time.

In addition to reducing the potential for accidental leaks to the environment, solidification will also reduce the volumes of wastes requiring storage by some 80 percent and will facilitate transport of these materials to a permanent disposal site. At the present time, DOE has not yet announced the location or design of the Federal repository to which the solidified wastes will eventually be shipped.

DOE is now evaporating its own high-level waste at the Hanford Reservation, converting part of it into solid salt cakes which are stored in underground tanks; and part into other solids which are stored in massive concrete structures above the ground. Similar practices are being followed at the Savannah River Plant. The DOE facility at INEL converts liquid high-level wastes to a granular solid form which is being stored in specially designed underground vaults (10).

Final plans for the ultimate disposal of the solidified high-level wastes have not been developed, and there are many complex factors which must be evaluated before a final selection is made. DOE is investigating several interim storage and ultimate disposal systems: underground storage or disposal in salt formations, caverns mined in bedrock, surface storage in massive concrete structures; and other more exotic concepts such as seabed, ice cap, or space disposal (11).

A major review of the technical alternative methods of managing radioactive wastes from the commercial nuclear reactor fuel cycle including the ultimate storage of this waste was conducted by the Energy Research and Development Administration (which was reorganized into DOE) in 1977. The review document provides a comprehensive compilation and description of alternative waste management operations, concepts, and characterizes the technologies in terms of state-or-readiness for use and development programs (12).

The treatment and disposal of radioactive wastes involve both environmental considerations and technical processing problems which are complex and potentially far-reaching because of the long effective half-lives of certain radionuclides. Moreover, the legal and administrative problems in radioactive waste management are more involved and difficult than in most general industrial operations.

REFERENCES

1. "Nuclear Power Fuel Reprocessing - Part I," page 61 December 28, 1959; "Hanford: Atoms for Peace - Part II," page 113, January 11, 1960; Idaho Falls: Atoms in the Desert - Part III," page 105, January 25, 1960; "Savannah River Does It Cheaper - Part IV," page 135, February 22, 1960; Oak Ridge: Home of The Bomb - Part V," page 137, March 7, 1960, Chemical Engineering.

2. G. W. Hogg, W. F. Holcomb, L. T. Lakey, L. H. Jones and D. D. Coward, A Survey of NRTS Wasts Management Practices, Volume I and II, USAEC Report ICP-1042-I & II, Idaho National Engineering Laboratory, Idaho Falls, Idaho, September 1971.

3. "Private Firm Gets Set to Reprocess Atomic Fuels," Chemical Engineering, page 68, April 29, 1963.

4. "NFS Quits Reprocessing, But Waste Solidification Problem Remains," Nucleonics Week, Vol. 17, No. 39, September 23, 1976.

5. "Nuclear Reprocessing at Standstill Despite Demand," Chemical Engineering, page 68, January 6, 1975.

6. L. J. Colby, Jr., "Fuel Reprocessing in the United States: A Review of Problems and Some Solutions," Nuclear News, page 68, January 1976.

7. "Nuclear-power prospects soured by oxide - fuel - reprocessing stall," Chemical Engineering, page 123, February 28, 1977.

8. Nuclear Fuel Reprocessing and High-Level Waste Management; Informational Hearings - Volume V, Reprocessing Part 2, California Energy Resources Conservation and Development Commission, Sacramento, California, March 8, 1977.

9. "Exxon Nuclear Tells Plan for Oak Ridge Facility," Nuclear News, page 59, March 1976.

10. W. L. Lennemann, "Management of Radioactive Aqueous Wastes from AEC Fuel-Reprocessing Operations," Nuclear Safety, Vol. 14, No. 5, page 482, September-October 1973.

11. High-Level Radioactive Waste Management Alternatives, USAEC Report WASH-1297, U. S. Atomic Energy Commission, Washington, D. C., May 1974.

12. Alternatives for Managing Wastes From Reactors and Past Fission Operations in The LWR Fuel Cycle, 5 Volumes, Report No. ERDA-76-43, U. S. Energy Research and Development Administration, Washington, D. C., May 1976.

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